THE INTERACTION OF RADIATION AND MATTER: QUANTUM THEORY

I. QUANTUM MECHANICS OF HARMONIC OSCILLATORS:[1]

To prepare the ground for the quantization of the electromagnetic field, let us revisit the **classical treatment** of a simple harmonic oscillator with one degree of freedom. We start, with the assertion that the Hamiltonian of such an oscillator must have the form.

$$\mathcal{H} = \frac{1}{2m}\left[p^2 + m^2\,\omega^2\,q^2\right]. \qquad \text{[I-1]}$$

where p and q are the cannonically conjugate variables. Consequently, the accordant Hamilton equations yield a pair of coupled, inhomogeneous, first-order equations -- *viz*.

$$\frac{dq}{dt} = \frac{\partial \mathcal{H}}{\partial p} = p/m \qquad \text{[I-2a]}$$

$$\frac{dp}{dt} = -\frac{\partial \mathcal{H}}{\partial q} = -m\,\omega^2\,q\ . \qquad \text{[I-2b]}$$

Combining these two equations, we see that formal classical analysis of the harmonic oscillator leads to a second-order homogeneous equation of motion

$$\frac{d^2q}{dt^2} = -\omega^2\,q^2 \qquad \text{[I-2c]}$$

with the evident complete solution

$$\begin{aligned} q(t) &= q(0)\,\cos\omega t + \frac{p(0)}{m\omega}\,\sin\omega t \\ p(t) &= -m\omega\,q(0)\,\sin\omega t + p(0)\,\cos\omega t \end{aligned} \qquad \text{[I-3]}$$

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[1] William Louisell's *Radiation and Noise in Quantum Electronics*, McGraw-Hill (1964) has an excellent set of pre-quantum optics reviews.

Equations [I-2a] and [I-2b] are **decoupled** by transforming to a new set of dynamic variables -- *viz.*[2]

$$\begin{aligned} a &= \sqrt{\frac{m}{2\omega}}\left[\omega q + i\ p/m\right] \\ a^\dagger &= \sqrt{\frac{m}{2\omega}}\left[\omega q - i\ p/m\right] \end{aligned} \qquad [\text{ I-4 }]$$

so that

$$\begin{aligned} q &= \frac{1}{\sqrt{2\,m\,\omega}}\left[a + a^\dagger\right] \\ p &= i\sqrt{\frac{m\,\omega}{2}}\left[a^\dagger - a\right] \end{aligned} \qquad [\text{ I-5 }]$$

In terms of these new variables the Hamiltonian becomes

$$\mathcal{H} = \omega\, a^\dagger a \qquad [\text{ I-6 }]$$

Thus, Equations [I-2a] and [I-2b] are transformed to the decoupled **normal mode** form -- *i.e.*

$$\begin{aligned} \frac{da}{dt} &= -i\,\omega\,a & \Leftrightarrow \qquad i\,\frac{da}{dt} &= \frac{\partial\mathcal{H}}{\partial a^\dagger} = \omega\,a \\ \frac{da^\dagger}{dt} &= i\,\omega\,a^\dagger & \Leftrightarrow \qquad i\,\frac{da^\dagger}{dt} &= -\frac{\partial\mathcal{H}}{\partial a} = -\omega\,a^\dagger \end{aligned} \qquad [\text{ I-7 }]$$

With this background, let us now turn to the quantum treatment of the harmonic oscillator. From the general theory discussed earlier, we make use of the ***classical analogy*** and postulate that the dynamic variables q and p become the Hermitian operators q and p which satisfy the commutation relations

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[2] In classical theory, the a and $a^\dagger$ variables are frequently referred to as normal mode amplitudes.

$$\begin{aligned} \left[q \, , q \right] &= 0 \\ \left[p \, , p \right] &= 0 \\ \left[q \, , p \right] &= i \hbar \end{aligned} \qquad \text{[I-8]}$$

The Schrödinger representation of the energy eigenvectors -- *i.e.* $u_n(q) \equiv \langle q | E_n \rangle$ -- satisfies the following representations of the energy eigenvalue equation

$$\langle \, q \, | \, \mathcal{H} \, | \, E_n \, \rangle = E_n \langle \, q | E_n \, \rangle \qquad \text{[I-9a]}$$

$$\langle \, q \, | \frac{1}{2\,m} \left[p^{\,2} + m^2 \omega^2 \, q^{\,2} \right] | \, E_n \, \rangle = E_n \langle \, q | E_n \, \rangle \qquad \text{[I-9b]}$$

$$\frac{1}{2\,m} \left[-\hbar^2 \frac{d^2}{dq^2} + m^2 \, \omega^2 \, q^2 \right] u_n(q) = E_n \, u_n(q) \quad . \qquad \text{[I-9c]}$$

The solution to this equation is familiar as

$$u_n(q) = \sqrt{\frac{1}{2^n \, n!} \sqrt{\frac{\omega}{\pi \, \hbar}}} \; H_n \left(\sqrt{\frac{m \, \omega}{\hbar}} \; q \right) \exp \left[-\frac{m \, \omega}{2 \, \hbar} \, q^2 \right] \qquad \text{[I-10a]}$$

where $H_n\left(\sqrt{m\,\omega / \hbar} \; q\right)$ is one of the Hermite polynomials discussed in the set of lecture notes entitled *On Classical Electromagnetic Fields* and the corresponding eigenvalues are given by

$$E_n = \hbar \, \omega_n = \hbar \, \omega \left(n + 1/2 \right) \; . \qquad \text{[I-10b]}$$

An alternative treatment of the harmonic oscillator provides a powerful set of new tools. Recall the transformation law between state vectors in the Schrödinger and Heisenberg pictures -- *viz*.

$$\left|\psi_S(t)\right\rangle = \mathcal{T}\,(t,0)\,\left|\psi_H(0)\right\rangle = \exp\left[-\frac{i\,\mathcal{H}\,t}{\hbar}\right]\left|\psi_H(0)\right\rangle \qquad [\text{ I-11 }]$$

which means that the operators transform according to the rules

$$\begin{aligned} q_H(t) &= \mathcal{T}^{\dagger}(t,0)\; q_S\; \mathcal{T}\,(t,0) \\ p_H(t) &= \mathcal{T}^{\dagger}(t,0)\; p_S\; \mathcal{T}\,(t,0) \end{aligned} \qquad [\text{ I-12 }]$$

The Heisenberg equations of motion for q_H and p_H are

$$\frac{dq_H}{dt} = \frac{\partial \mathcal{H}_H}{\partial p_H} = p_H/m \qquad [\text{ I-13a }]$$

$$\frac{dp_H}{dt} = -\frac{\partial \mathcal{H}_H}{\partial q_H} = -m\omega^2\, q_H \qquad [\text{ I-13b }]$$

By analogy with Equation [I-3], we see that the time evolution

$$\begin{aligned} q_H(t) &= \mathcal{T}^{\dagger}(t,0)\; q_S\; \mathcal{T}\,(t,0) = q_S\,\cos\omega t + \frac{p_S}{m\omega}\sin\omega t \\ p_H(t) &= \mathcal{T}^{\dagger}(t,0)\; p_S\; \mathcal{T}\,(t,0) = -m\omega\, q_S\,\sin\omega t + p_S\,\cos\omega t \end{aligned} \qquad [\text{ I-14 }]$$

is consistent with Equations [I-13a] and [I-13b]. By analogy with Equation [I-4] we introduce two critically important (non-Hermitian) operators -- *i.e.*

$$\begin{aligned} a &= \sqrt{\frac{m}{2\hbar\omega}}\left[\omega\, q + i\, p/m\right] \\ a^{\dagger} &= \sqrt{\frac{m}{2\hbar\omega}}\left[\omega\, q - i\, p/m\right] \end{aligned} \qquad [\text{ I-15 }]$$

or

$$q = \sqrt{\frac{\hbar}{2 m \omega}} \left[a + a^{\dagger} \right] \qquad p = i \sqrt{\frac{m \hbar \omega}{2}} \left[a^{\dagger} - a \right] \qquad \text{[I-16]}$$

with the commutation relation

$$\left[a, a^{\dagger} \right] = 1 \qquad \text{[I-17]}$$

Thus

$$\mathcal{H} = \frac{\hbar\omega}{2} \left[a a^{\dagger} + a^{\dagger} a \right] = \hbar\omega \left[a^{\dagger} a + 1/2 \right] = \hbar\omega \left[\mathcal{N} + 1/2 \right] \qquad \text{[I-18]}$$

where $\mathcal{N} = a^{\dagger} a$ is an important (as yet uninterpreted) Hermitian operator (observable). The Heisenberg equations of motion become

$$\frac{d a_H}{dt} = \frac{1}{i \hbar} \left[a_H, \mathcal{H}_H \right] = -i \omega a_H \qquad \frac{d a_H^{\dagger}}{dt} = \frac{1}{i \hbar} \left[a_H^{\dagger}, \mathcal{H}_H \right] = +i \omega a_H^{\dagger} \qquad \text{[I-19]}$$

with the solutions

$$a_H(t) = \mathcal{T}^{\dagger}(t,0)\, a_S\, \mathcal{T}(t,0) = a_S \exp(-i \omega t) \qquad a_H^{\dagger}(t) = \mathcal{T}^{\dagger}(t,0)\, a_S^{\dagger}\, \mathcal{T}(t,0) = a_S^{\dagger} \exp(+i \omega t) \qquad \text{[I-20]}$$

Consider the following equivalent eigenvalues problems -- *viz.*[3]

$$\mathcal{H} \left| E' \right\rangle = E' \left| E' \right\rangle \quad \text{and} \quad \mathcal{N} \left| n' \right\rangle = n' \left| n' \right\rangle \qquad \text{[I-21]}$$

First note that

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[3] It follows from Equation [I-18] that $E' = \hbar\omega \left[n' + 1/2 \right]$.

$$\begin{aligned} \left[\mathcal{N}, a\right] &= \mathcal{N}\, a - a\, \mathcal{N} = a^{\dagger} a\, a - a\, a^{\dagger} a = \left[a^{\dagger}, a\right] a = -a \\ \left[\mathcal{N}, a^{\dagger}\right] &= \mathcal{N}\, a^{\dagger} - a^{\dagger} \mathcal{N} = a^{\dagger} a\, a^{\dagger} - a^{\dagger} a^{\dagger} a = a^{\dagger}\left[a, a^{\dagger}\right] = a^{\dagger} \end{aligned} \qquad [\text{ I-22a }]$$

or

$$\begin{aligned} \mathcal{N}\, a &= a\left(\mathcal{N} - 1\right) \\ \mathcal{N}\, a^{\dagger} &= a^{\dagger}\left(\mathcal{N} + 1\right) \end{aligned} \qquad [\text{ I-22b }]$$

Thus, we see that

$$\begin{aligned} \mathcal{N}\left\{a\,|n'\rangle\right\} &= a\,\left(\mathcal{N} - 1\right)|n'\rangle = \left(n' - 1\right) a\,|n'\rangle = \left(n' - 1\right)|n' - 1\rangle \\ \mathcal{N}\left\{a^{\dagger}|n'\rangle\right\} &= a^{\dagger}\left(\mathcal{N} + 1\right)|n'\rangle = \left(n' + 1\right) a^{\dagger}|n'\rangle = \left(n' + 1\right)|n' + 1\rangle\, . \end{aligned} \qquad [\text{ I-23 }]$$

That is, if $|n'\rangle$ is an eigenket of the operator $\mathcal{N}$ with eigenvalue n', then $a\,|n'\rangle$ is also an eigenket, but with the eigenvalue $n' - 1$. Similarly, if $|n'\rangle$ is an eigenket of the operator $\mathcal{N}$ with eigenvalue n', then $a^{\dagger}|n'\rangle$ is an eigenvector with eigenvalue $\left(n' + 1\right)$. Hence, the operators a and $a^{\dagger}$ "generate" new eigenkets which have associated eigenvalues differing by one from eigenvalues associated with the original eigenkets. It follows that each eigenvalue of $\mathcal{N}$ differs from all other eigenvalues by an integer!

Normalization of these eigenkets proceeds as follows:

Suppose

$$a\,|n\rangle = c_n\,|n - 1\rangle \qquad [\text{ I-24 }]$$

Then

$$\begin{aligned} n\langle n|n\rangle &= \langle n|\mathcal{N}\,|n\rangle = \langle n|a^{\dagger}\,a\,|n\rangle = c_n\langle n|\,a^{\dagger}|n - 1\rangle \\ &= c_n\langle n - 1|\,a\,|n\rangle = |c_n|^2\langle n - 1|n - 1\rangle \end{aligned} \qquad [\text{ I-25 }]$$

but we require $\langle n|n\rangle = \langle n-1|n-1\rangle = 1$ so that $c_n = \sqrt{n}$. Therefore

$$\begin{aligned} \mathcal{N}\,|n\rangle &= n\,|n\rangle \\ a\,|0\rangle &= 0 \\ a\,|n\rangle &= \sqrt{n}\,|n-1\rangle \\ a^{\dagger}|n\rangle &= \sqrt{n+1}\,|n+1\rangle \end{aligned} \qquad [\text{ I-26 }]$$

which leads to the powerful **state vector generating rule** -- *viz.*

$$|n\rangle = \frac{\left(a^{\dagger}\right)^{n}}{\sqrt{n!}}|0\rangle \qquad [\text{ I-27 }]$$

There is another interest connect between the two alternative treatments of a harmonic oscillator: we may use Equations [I-26] and [I-27] to obtain the coordinate representatives of the harmonic oscillator states. Using Equation [I-15] we see that

$$\begin{aligned} \langle q|a\,|0\rangle &= \sqrt{\frac{m}{2\,\hbar\omega}}\langle q|\left[\omega\,q + i\,p\,/m\right]|0\rangle \\ &= \sqrt{\frac{m}{2\,\hbar\omega}}\left[\omega\,q + \frac{\hbar}{m}\frac{d}{dq}\right]\langle q|0\rangle = 0 \end{aligned} \qquad [\text{ I-28 }]$$

Therefore

$$\langle q|0\rangle = c_0\,\exp\left[-\frac{m\omega}{2\hbar}\,q^2\right] \qquad [\text{ I-29 }]$$

and with the equivalence

$$a^{\dagger} \Leftrightarrow \sqrt{\frac{m}{2\,\hbar\,\omega}}\left[\omega\,q - \frac{\hbar}{m}\frac{d}{dq}\right] \qquad [\text{ I-30 }]$$

Equation [I-27 } yields

$$\langle q|n\rangle = \frac{1}{\sqrt{n!}}\left[\sqrt{\frac{m}{2\,\hbar\omega}}\right]^n \left[\omega\, q - \frac{\hbar}{m}\frac{d}{dq}\right]^n \langle q|0\rangle \qquad [\text{ I-31 }]$$

which is the generator of the eigenfunction displayed in Equation [I-10a].

The connection between bosons and harmonic oscillators:

There are two distinct interpretations of the energy formula $\mathcal{E}_n = \hbar\omega\left(n + \tfrac{1}{2}\right)$:

a. The formula describes the energy eigenvalue which corresponds to a given eigenstate of a particular harmonic oscillator.

b. The formula describes the total energy of a system of n identical noninteracting quanta (particles), each of which is in the same dynamic state. The energy of the particle state is $\hbar\omega$.

Again we turn to Dirac[4] who rigorously articulates the equivalence of these two view points. To quote his conclusion "……**This means that the dynamic system consisting of an assembly of similar bosons is equivalent to the dynamic system consisting of a set of oscillators - the two systems are just the same system looked at from two different points of view. There is one oscillator associated with each independent boson state. We have here one of the most fundamental results of quantum mechanics, which enables a unification of the wave and corpuscular theories of light to be effected**………**Thus a set of harmonic oscillators is equivalent to an assembly of bosons in stationary states with no interactions between**

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[4] In Sections 60 and 61 of P. A. M. Dirac's *The Principles of Quantum Mechanics* (Revised fourth edition), Oxford University Press (1967).

them. If an oscillator of the set is in its n'th quantum state, there are n' bosons in the associated boson state."

II. CANONICAL QUANTIZATION OF ELECTRODYNAMICS:

With the foregoing preparation, we are now in a position to apply the *classical analogy* or *canonical quantization* program to achieve the ***second quantization*** of the electromagnetic field.[5] As our starting point and for reference, we, once again, set forth the vacuum or microscopic Maxwell's equations in the time domain:

$$\vec{\nabla}\times\vec{\mathbf{E}}\left(\vec{\mathbf{r}},t\right)=-\frac{\partial}{\partial t}\vec{\mathbf{B}}\left(\vec{\mathbf{r}},t\right) \qquad [\text{ II-1a }]$$

$$\vec{\nabla}\times\vec{\mathbf{B}}\left(\vec{\mathbf{r}},t\right)=\mu_0\,\vec{\mathbf{J}}\left(\vec{\mathbf{r}},t\right)+\varepsilon_0\mu_0\frac{\partial}{\partial t}\vec{\mathbf{E}}\left(\vec{\mathbf{r}},t\right) \qquad [\text{ II-1b }]$$

$$\vec{\nabla}\cdot\vec{\mathbf{E}}\left(\vec{\mathbf{r}},t\right)=\rho\left(\vec{\mathbf{r}},t\right)/\varepsilon_0 \qquad [\text{ II-1c }]$$

$$\vec{\nabla}\cdot\vec{\mathbf{B}}\left(\vec{\mathbf{r}},t\right)=0 \qquad [\text{ II-1d }]$$

The canonical formulation of classical electrodynamics (**Jeans' Theorem**) is most conveniently achieved in terms of the (magnetic) vector potential in the time domain -- *viz*.

$$\vec{\mathbf{B}}\left(\vec{\mathbf{r}},t\right)=\vec{\nabla}\times\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right) \qquad [\text{ II-2a }]$$

$$\vec{\mathbf{E}}\left(\vec{\mathbf{r}},t\right)=-\frac{\partial}{\partial t}\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)-\vec{\nabla}\phi\left(\vec{\mathbf{r}},t\right) \qquad [\text{ II-2b }]$$

so that

$$\vec{\nabla}\left[\vec{\nabla}\cdot\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)\right]-\nabla^2\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)+\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)+\frac{1}{c^2}\frac{\partial}{\partial t}\vec{\nabla}\phi\left(\vec{\mathbf{r}},t\right)=\mu_0\,\vec{\mathbf{J}}\left(\vec{\mathbf{r}},t\right) \qquad [\text{ II-3a }]$$

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[5] In common usage, the process of treating the cordinates q_i and p_i as quantized variables is called *first quantization*. *Second quantization* is the process of quantizing ***fields*** -- say, $\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)$ -- which have an infinite number of dequees of freedom.

$$-\varepsilon_0 \vec{\ }\ \frac{\partial}{\partial t}\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)-\varepsilon_0\ {}^2\phi\left(\vec{\mathbf{r}},t\right)=\rho\left(\vec{\mathbf{r}},t\right)w \qquad [\text{ II-3b }]$$

In QED (Quantum Electrodynamics) it is *convenient and traditional* to make use of the **Coulomb gauge --** *i.e.* $\vec{\ }\ \vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)=0$ -- so that

$${}^2\,\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)=-\mu_0\,\vec{\mathbf{J}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right) \qquad [\text{ II-4a }]$$

$${}^2\,\phi\left(\vec{\mathbf{r}},t\right)=-\rho\left(\vec{\mathbf{r}},t\right)/\varepsilon_0 \qquad [\text{ II-4b }]$$

where $\vec{\mathbf{J}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right)=\vec{\mathbf{J}}\left(\vec{\mathbf{r}},t\right)-\vec{\mathbf{J}}_{\mathrm{L}}\left(\vec{\mathbf{r}},t\right)=\vec{\mathbf{J}}\left(\vec{\mathbf{r}},t\right)-\varepsilon_0\frac{\partial}{\partial t}\ \phi\left(\vec{\mathbf{r}},t\right)$ is the so called ***transverse*** current density. Since $\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)$ is completely determined by the transverse current density in the Coulomb gauge, electromagnetic problems become in a sense separable -- *i.e.*

The *transverse* field problem:

$$\begin{aligned}
&\vec{\ }\ \vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right)=0\\
&\vec{\ }\times\vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right)=-\mu_0\frac{\partial}{\partial t}\vec{\mathbf{H}}\left(\vec{\mathbf{r}},t\right) \qquad [\text{ II-5a }]\\
&\vec{\ }\times\vec{\mathbf{H}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right)=\vec{\mathbf{J}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right)+\frac{1}{c^2}\frac{\partial}{\partial t}\vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right)
\end{aligned}$$

The *longitudinal* field problem:

$$\begin{aligned}
&\vec{\ }\ \vec{\mathbf{E}}_{\mathrm{L}}\left(\vec{\mathbf{r}},t\right)=\rho\left(\vec{\mathbf{r}},t\right)/\varepsilon_0\\
&\vec{\mathbf{J}}_{\mathrm{L}}\left(\vec{\mathbf{r}},t\right)=-\varepsilon_0\frac{\partial}{\partial t}\vec{\mathbf{E}}_{\mathrm{L}}\left(\vec{\mathbf{r}},t\right) \qquad [\text{ II-5b }]
\end{aligned}$$

We turn now explicitly to a treatment of the ***free electromagnetic field*** -- formally the case of $\vec{\mathbf{J}}_T(\vec{\mathbf{r}},t) \equiv 0$ wherein

$$\nabla^2 \vec{\mathbf{A}}(\vec{\mathbf{r}},t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{\mathbf{A}}(\vec{\mathbf{r}},t) = 0 \qquad [\text{ II-6 }]$$

We look for solutions in the form

$$\vec{\mathbf{A}}(\vec{\mathbf{r}},t) = \frac{1}{2\sqrt{\varepsilon_0}}\sum_s\left\{\alpha_s(t)\,\vec{\mathbf{u}}_s(\vec{\mathbf{r}}) + c.c.\right\} \qquad [\text{ II-7 }]$$

Substituting into Equation [II-6] we obtain

$$\sum_s\left\{\alpha_s(t)\,\vec{\mathbf{u}}_s(\vec{\mathbf{r}})\left[\frac{\nabla^2\vec{\mathbf{u}}_s(\vec{\mathbf{r}})}{\vec{\mathbf{u}}_s(\vec{\mathbf{r}})} - \frac{1}{c^2}\frac{\ddot{\alpha}_s(t)}{\alpha_s(t)}\right] + c.c.\right\} = 0 \qquad [\text{ II-8 }]$$

Thus we may apply **separation of variables** techniques and the original problem is divided in to **two new and distinct** problems -- *viz.* solutions of the following set of equations:

$$\nabla^2\vec{\mathbf{u}}_s(\vec{\mathbf{r}}) + \frac{\omega_s^2}{c^2}\vec{\mathbf{u}}_s(\vec{\mathbf{r}}) = 0 \qquad [\text{ II-9 }]$$

$$\ddot{\alpha}_s(t) + \omega_s^2\,\alpha_s(t) = 0 \qquad [\text{ II-10 }]$$

where the ω_s's are **separation constants**. The spatial equations allow us to treat the boundary value problem of the cavity or defining field space in whatever detail that might seem appropriate in a particular case.[6] But the *bottom line* is that the boundary condition taken together with the characteristics of the normal modes or eigenfunctions determine the ω_s's which may, in turn, be identified as the eigenfrequencies of the normal modes.

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[6] The $\vec{\mathbf{u}}_i(\vec{\mathbf{r}})$'s do not form a true complete set of solutions since no longitudinal vector field can be expanded in terms of such *divergentless* functions.

Thus, we may now write

$$\begin{aligned}\vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}},t) = -\frac{\partial}{\partial t}\vec{\mathbf{A}}\,(\vec{\mathbf{r}},t) &= -\frac{1}{2\sqrt{\varepsilon_0}}\sum_{s}\left\{\dot{\alpha}_s(t)\,\vec{\mathbf{u}}_s(\vec{\mathbf{r}}) + c.c.\right\} \\ &= \frac{1}{2\sqrt{\varepsilon_0}}\sum_{s}\left\{i\,\omega_s\alpha_s(t)\,\vec{\mathbf{u}}_s(\vec{\mathbf{r}}) + c.c.\right\}\end{aligned} \qquad [\text{ II-11 }]$$

and

$$\vec{\mathbf{H}}_{\mathrm{T}}(\vec{\mathbf{r}},t) = \frac{1}{\mu_0}\,\vec{\nabla}\times\vec{\mathbf{A}}\,(\vec{\mathbf{r}},t) = \frac{c}{2\sqrt{\mu_0}}\sum_{s}\left\{\alpha_s(t)\,\vec{\nabla}\times\vec{\mathbf{u}}_s(\vec{\mathbf{r}}) + c.c.\right\}. \qquad [\text{ II-12 }]$$

The crucial step required in establishing Jeans' Theorem is the expansion of the instantaneous value of the stored electromagnetic energy in terms of the cavity modes -- *viz.*

$$\langle W(t)\rangle = \frac{1}{2V}\iiint\left[\varepsilon_0\left|\vec{\mathbf{E}}(\vec{\mathbf{r}},t)\right|^2 + \mu_0\left|\vec{\mathbf{H}}(\vec{\mathbf{r}},t)\right|^2\right]dV \qquad [\text{ II-13a }]$$

which, in light of Equations [II-11] and [II-12], becomes

$$\langle W(t)\rangle = \frac{1}{4V}\sum_{s,s'}\iiint\left[\omega_s\omega_{s'}\,\vec{\mathbf{u}}_s\cdot\vec{\mathbf{u}}^{*}_{s'} + c^2\,\vec{\nabla}\times\vec{\mathbf{u}}_s\cdot\vec{\nabla}\times\vec{\mathbf{u}}^{*}_{s'}\right]\alpha_s(t)\,\alpha^{*}_{s'}(t)\,dV \qquad [\text{ II-13b }]$$

To proceed we need the value of the integral $\iiint\limits_{\text{cavity}} \left[\vec{\nabla} \times \vec{\mathbf{u}}_{\mathrm{s}} \cdot \vec{\nabla} \times \vec{\mathbf{u}}^{*}_{\mathrm{s}'} \right] dV$. Using a well known vector identity,[7] we obtain

$$\iiint \left[\vec{\nabla} \times \vec{\mathbf{u}}_{\mathrm{s}} \cdot \vec{\nabla} \times \vec{\mathbf{u}}^{*}_{\mathrm{s}'} \right] dV = \iint \vec{\mathbf{u}}_{\mathrm{s}} \times \left(\vec{\nabla} \times \vec{\mathbf{u}}^{*}_{\mathrm{s}'} \right) \cdot d\vec{S} + \iiint \vec{\mathbf{u}}_{\mathrm{s}} \cdot \vec{\nabla} \times \left(\vec{\nabla} \times \vec{\mathbf{u}}^{*}_{\mathrm{s}'} \right) dV \qquad \text{[II-14a]}$$

By boundary value arguments we may easily show that the first term on the RHS of this equation vanishes and by using an even more familiar (famous) vector identity[8] for a divergenceles field, we obtain

$$\iiint \left[\vec{\nabla} \times \vec{\mathbf{u}}_{\mathrm{s}} \cdot \vec{\nabla} \times \vec{\mathbf{u}}^{*}_{\mathrm{s}'} \right] dV = -\iiint \vec{\mathbf{u}}_{\mathrm{s}} \cdot \nabla^{2} \vec{\mathbf{u}}^{*}_{\mathrm{s}'}\, dV = \frac{\omega^{2}_{\mathrm{s}'}}{c^{2}} \iiint \vec{\mathbf{u}}_{\mathrm{s}} \cdot \vec{\mathbf{u}}^{*}_{\mathrm{s}'}\, dV \qquad \text{[II-14b]}$$

Therefore, Equation [II-13b] becomes

$$\langle W(t) \rangle = \frac{1}{4V} \sum_{\mathrm{s},\mathrm{s}'} \left[\omega_{\mathrm{s}} \omega_{\mathrm{s}'} + \omega^{2}_{\mathrm{s}'} \right] \alpha_{\mathrm{s}}(t)\, \alpha^{*}_{\mathrm{s}'}(t) \iiint \vec{\mathbf{u}}_{\mathrm{s}} \cdot \vec{\mathbf{u}}^{*}_{\mathrm{s}'}\, dV \qquad \text{[II-15]}$$

From Equation [II-14b] we may also write

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[7] Namely, that $\vec{\nabla} \cdot \left(\vec{\mathbf{X}} \times \vec{\nabla} \times \vec{\mathbf{Y}} \right) = \left(\vec{\nabla} \times \vec{\mathbf{X}} \right) \cdot \left(\vec{\nabla} \times \vec{\mathbf{Y}} \right) - \vec{\mathbf{X}} \cdot \left(\vec{\nabla} \times \vec{\nabla} \times \vec{\mathbf{Y}} \right)$

[8] Namely, that

$$\begin{aligned} \vec{\nabla} \cdot \left[\vec{\mathbf{X}} \times \left(\vec{\nabla} \times \vec{\mathbf{Y}} \right) \right] &= \left(\vec{\nabla} \times \vec{\mathbf{X}} \right) \cdot \left(\vec{\nabla} \times \vec{\mathbf{Y}} \right) - \vec{\mathbf{X}} \cdot \vec{\nabla} \times \left(\vec{\nabla} \times \vec{\mathbf{Y}} \right) \\ &= \left(\vec{\nabla} \times \vec{\mathbf{X}} \right) \cdot \left(\vec{\nabla} \times \vec{\mathbf{Y}} \right) - \vec{\mathbf{X}} \cdot \vec{\nabla} \left(\vec{\nabla} \cdot \vec{\mathbf{Y}} \right) + \vec{\mathbf{X}} \cdot \nabla^{2} \vec{\mathbf{Y}} \end{aligned}$$

$$\iiint \vec{\;} \times \left[\vec{\mathbf{u}}_{s} \times \vec{\;} \times \vec{\mathbf{u}}^{*}_{s'} - \vec{\mathbf{u}}^{*}_{s'} \times \vec{\;} \times \vec{\mathbf{u}}_{s} \right] dV = \frac{1}{c^{2}} \left(\omega^{2}_{s'} - \omega^{2}_{s} \right) \iiint \vec{\mathbf{u}}_{s} \;\; \vec{\mathbf{u}}^{*}_{s'} \, dV \quad \text{II-16a]}$$

and, again, by boundary value arguments we may easily show that the resultant surface imtegral on the left

$$\left(\omega^{2}_{s'} - \omega^{2}_{s} \right) \iiint \vec{\mathbf{u}}_{s} \;\; \vec{\mathbf{u}}^{*}_{s'} \, dV = 0 \qquad \text{[II-16b]}$$

Therefore, we may take

$$\iiint \left[\vec{\mathbf{u}}_{s} \;\; \vec{\mathbf{u}}^{*}_{s'} \right] dV = \delta_{ss'} \qquad \text{[II-17]}$$

so that Equation [II-15] becomes

$$\langle W(t) \rangle = \frac{1}{2} \sum_{s} \omega^{2}_{s} \, \alpha_{s} \, \alpha^{*}_{s} \qquad \text{[II-18]}$$

which when compared to Equation [I-6][9] **is effectively the content of Jean's Theorem** with

$$\begin{aligned} \sqrt{\frac{\omega_{s}}{2}}\, \alpha_{s} \qquad & a = \sqrt{\frac{m}{2\,\omega}} \left[\omega\, q + i\, p/m \right] \\ \sqrt{\frac{\omega_{s}}{2}}\, \alpha^{*}_{s} \qquad & a^{\dagger} = \sqrt{\frac{m}{2\,\omega}} \left[\omega\, q - i\, p/m \right] \end{aligned} \qquad \text{[II-19]}$$

---

[9] That is $\mathcal{H} = \omega\, a^{\dagger} a$

To accomplish the *canonical quantization* program, field variables are expressed as field operators by making the identification

$$\sqrt{\frac{\omega_s}{2}}\,\alpha_s \qquad \sqrt{\frac{m}{2\,\omega}}\left[\omega\, q + i\, p\,/m\right] = \sqrt{\hbar}\, a$$
$$\sqrt{\frac{\omega_s}{2}}\,\alpha^*_s \qquad \sqrt{\frac{m}{2\,\hbar\omega}}\left[\omega\, q - i\, p\,/m\right] = \sqrt{\hbar}\, a^{\dagger} \qquad [\text{ II-20 }]$$

which leads to the following set of field operators:

$$\mathcal{H} = \sum_s \frac{\hbar\omega_s}{2}\left\{a_s\, a_s^{\dagger} + a_s^{\dagger}\, a_s\right\}$$

$$\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right) = \sum_s \sqrt{\frac{\hbar}{2\,\omega_s\,\varepsilon_0}}\left\{a_s(t)\,\vec{\mathbf{u}}_s\left(\vec{\mathbf{r}}\right) + a_s^{\dagger}(t)\,\vec{\mathbf{u}}^*_{s'}\right\}$$

$$\vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right) = \sum_s \sqrt{\frac{\hbar\omega_s}{2\varepsilon_0}}\left\{a_s(t)\,\vec{\mathbf{u}}_s\left(\vec{\mathbf{r}}\right) - a_s^{\dagger}(t)\,\vec{\mathbf{u}}^*_{s'}\right\}$$

$$[\text{ II-21 }]$$

$$\vec{\mathbf{H}}_{\mathrm{T}}\left(\vec{\mathbf{r}},t\right) = c\sum_s \sqrt{\frac{\hbar}{2\,\omega_s\,\mu_0}}\left\{a_s(t)\;\vec{\ }\times\vec{\mathbf{u}}_s\left(\vec{\mathbf{r}}\right) + a_s^{\dagger}(t)\;\vec{\ }\times\vec{\mathbf{u}}^*_{s'}\right\}$$

The Plane Wave Expansion of the Electromagnetic Field Hamiltonian:

To be definite, we may write an explicit plane wave representation for the field as

$$\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right) = \sum_s\sum_{\sigma=1}^{2}\sqrt{\frac{\hbar}{2\omega_s\,\varepsilon_0\,V}}\;\hat{\mathbf{e}}_{i\,\sigma}\left\{a_{s\sigma}\exp\left[i\left(\vec{\mathbf{k}}_s\;\;\vec{\mathbf{r}}-\omega_s\,t\right)\right] + a^{\dagger}_{s\sigma}\exp\left[-i\left(\vec{\mathbf{k}}_s\;\;\vec{\mathbf{r}}-\omega_s\,t\right)\right]\right\} \qquad [\text{ II-22a }]$$

or[10]

$$\vec{\mathbf{A}}\left(\vec{\mathbf{r}},t\right)=\sum_{s}\sum_{\sigma=1}^{2}\sqrt{\frac{\hbar}{2\omega_{s}\,\varepsilon_{0}}}\left\{a_{s\sigma}(t)\,\vec{\mathbf{u}}_{s\sigma}(\vec{\mathbf{r}})+a_{s\sigma}^{\dagger}(t)\,\vec{\mathbf{u}}_{s\sigma}(\vec{\mathbf{r}})\right\} \qquad \text{[II-22b]}$$

where

$$\vec{\mathbf{u}}_{s\sigma}(\vec{\mathbf{r}})=\frac{1}{\sqrt{V}}\,\hat{\mathbf{e}}_{s\sigma}\,\exp\left[i\,\vec{\mathbf{k}}_{s}\cdot\vec{\mathbf{r}}\right] \quad \text{and} \quad \vec{\mathbf{u}}_{s\sigma}(\vec{\mathbf{r}})=\frac{1}{\sqrt{V}}\,\hat{\mathbf{e}}_{s\sigma}\,\exp\left[-i\,\vec{\mathbf{k}}_{s}\cdot\vec{\mathbf{r}}\right] \qquad \text{[II-23a]}$$

$$a_{s\sigma}(t)=a_{s\sigma}(0)\,\exp\left[-i\,\omega_{s}\,t\right] \quad \text{and} \quad a_{s\sigma}^{\dagger}(t)=a_{s\sigma}^{\dagger}(0)\,\exp\left[+i\,\omega_{s}\,t\right] \qquad \text{[II-23b]}$$

Of course, $k_s=\left|\vec{\mathbf{k}}_s\right|=\omega_s/c$ in all of these expansions. Further the electric field expansion is given by

$$\begin{aligned}\vec{\mathbf{E}}(\vec{\mathbf{r}},t)&=i\sum_{s}\sum_{\sigma=1}^{2}\sqrt{\frac{\hbar\,\omega_{s}}{2\,\varepsilon_{0}}}\left\{a_{s\sigma}(t)\,\vec{\mathbf{u}}_{s\sigma}(\vec{\mathbf{r}})-a_{s\sigma}^{\dagger}(t)\,\vec{\mathbf{u}}_{s\sigma}(\vec{\mathbf{r}})\right\}\\&=i\sum_{s}\sum_{\sigma=1}^{2}\hat{\mathbf{e}}_{s\sigma}\,\mathcal{E}_{s}\left\{a_{s\sigma}\,\exp\left[i\,\vec{\mathbf{k}}_{s}\cdot\vec{\mathbf{r}}-i\,\omega_{s}\,t\right]-a_{s\sigma}^{\dagger}(t)\,\exp\left[i\,\vec{\mathbf{k}}_{s}\cdot\vec{\mathbf{r}}-i\,\omega_{s}\,t\right]\right\}\end{aligned} \qquad \text{[II-24a]}$$

where $\mathcal{E}_s=\sqrt{\dfrac{\hbar\omega_s}{2\varepsilon_0\,V}}$ and the magnetic field expansion by

$$\vec{\mathbf{H}}(\vec{\mathbf{r}},t)=\frac{i}{c\,\mu_{0}}\sum_{s}\sum_{\sigma=1}^{2}\sqrt{\frac{\hbar\omega_{s}}{2\varepsilon_{0}}}\left\{a_{s\sigma}(t)\left[\hat{\mathbf{k}}_{s}\times\vec{\mathbf{u}}_{s\sigma}(\vec{\mathbf{r}})\right]-a_{s\sigma}^{\dagger}(t)\left[\hat{\mathbf{k}}_{s}\times\vec{\mathbf{u}}_{s\sigma}(\vec{\mathbf{r}})\right]\right\} \qquad \text{[II-24b]}$$

---

[10] This expansion is often written as $\vec{\mathbf{E}}(\vec{\mathbf{r}},t)=\vec{\mathbf{E}}^{(+)}(\vec{\mathbf{r}},t)+\vec{\mathbf{E}}^{(-)}(\vec{\mathbf{r}},t)$ where

$$\vec{\mathbf{E}}^{(+)}(\vec{\mathbf{r}},t)=i\sum_{s}\sum_{\sigma=1}^{2}\hat{\mathbf{e}}_{s\sigma}\,\mathcal{E}_{s}\,a_{s\sigma}\,\exp\left[i\,\vec{\mathbf{k}}_{s}\cdot\vec{\mathbf{r}}-i\,\omega_{s}\,t\right] \quad \text{and} \quad \vec{\mathbf{E}}^{(-)}(\vec{\mathbf{r}},t)=-i\sum_{s}\sum_{\sigma=1}^{2}\hat{\mathbf{e}}_{s\sigma}\,\mathcal{E}_{s}\,a_{s\sigma}^{\dagger}(t)\,\exp\left[i\,\vec{\mathbf{k}}_{s}\cdot\vec{\mathbf{r}}-i\,\omega_{s}\right.$$

In light of Equations [II-18] and [II-21], the Hamiltonian of the radiation field is

$$\mathcal{H}_{rad} = \frac{1}{2}\sum_{\{l\}}\sum_{\sigma=1}^{2}\hbar\,\omega_{\{l\}}\left\{a_{\{l\}\sigma}\,a^{\dagger}_{\{l\}\sigma} + a^{\dagger}_{\{l\}\sigma}\,a_{\{l\}\sigma}\right\} = \sum_{\{l\}}\sum_{\sigma=1}^{2}\hbar\,\omega_{\{l\}}\left\{\mathcal{N}_{\{l\}\sigma} + 1/2\right\} \quad [\text{ II-25 }]$$

where

$$\begin{aligned}
\sum_{\{l\}} &\equiv \sum_{l_x=-\infty}^{+\infty}\sum_{l_y=-\infty}^{+\infty}\sum_{l_z=-\infty}^{+\infty} \\
\vec{\mathbf{k}}_{\{\}} &\equiv \frac{2\pi}{L}\left[l_x\,\hat{\mathbf{x}} + l_y\hat{\mathbf{y}} + l_z\hat{\mathbf{z}}\right] \qquad [\text{ II-26 }] \\
\omega_{\{\}} &\equiv c\left|\,\vec{\mathbf{k}}_{\{l\}}\,\right|
\end{aligned}$$

The electromagnetic momentum (Poynting vector divided by c^2) is given classical by

$$\vec{\mathcal{M}} = \frac{1}{c^2}\iiint_{\text{cavity}}\vec{\mathbf{E}}\times\vec{\mathbf{H}}\,dV \qquad [\text{ II-27a }]$$

and in terms of the second quantization operators it becomes

$$\begin{aligned}
\vec{\mathcal{M}} &= \frac{1}{2}\sum_{\{l\}}\sum_{\sigma=1}^{2}\hbar\,\vec{\mathbf{k}}_{\{l\}}\left\{a_{\{l\}\sigma}\,a^{\dagger}_{\{l\}\sigma} + a_{\{l\}\sigma}\,a^{\dagger}_{\{l\}\sigma}\right\} \qquad [\text{ II-27b }] \\
&= \sum_{\{l\}}\sum_{\sigma=1}^{2}\hbar\,\vec{\mathbf{k}}_{\{l\}}\,\mathcal{N}
\end{aligned}$$

Thus, the Fock or number states are eigenstates of both the energy and the momentum of the field.

III. REPRESENTATIONS OF PHOTON STATES

1. Fock or "Number" States: .[11]

As we have seen, the Fock or *number* states

$$\left|\left\{n_{\vec{\mathbf{k}}}\right\}\right\rangle \equiv \prod_{s\,\sigma}\left|n_{\vec{\mathbf{k}}_s\,\sigma}\right\rangle \qquad [\text{ III-1 }]$$

are complete set eigenstates of an important group of commuting observables -- *viz*. $\mathcal{H}_{rad}$, $\mathcal{N}$ and $\vec{\mathcal{M}}$.

Reprise of Characteristics and Properties of Fock States:

a. **The expectation value of the number operator and the *fractional uncertainty* associated with a single Fock state**:

$$\langle n|\mathcal{N}\;|n\rangle = n \qquad [\text{ III-2a }]$$

$$\Delta n = [\text{ "uncertainty" }] = \sqrt{\left\{\langle n|\mathcal{N}^{\,2}\,|n\rangle - \langle n|\mathcal{N}\;|n\rangle^2\right\}} = 0 \qquad [\text{ III-2b }]$$

b. **Expectation value of the fields associated with a single mode**:

For one mode Equations [II-24a] and [II-24b] reduce to

$$\vec{\mathbf{E}}\left(\vec{\mathbf{r}},t\right) = i\,\hat{\mathbf{e}}\,\mathcal{E}\left\{a\;\exp\left[i\,\vec{\mathbf{k}}\;\;\vec{\mathbf{r}} - i\,\omega\,t\right] - a^{\dagger}\exp\left[-i\,\vec{\mathbf{k}}\;\;\vec{\mathbf{r}} - i\,\omega\,t\right]\right\} \qquad [\text{ III-3a }]$$

$$\vec{\mathbf{H}}\left(\vec{\mathbf{r}},t\right) = i\sqrt{\frac{\varepsilon_0}{\mu_0}}\;\mathcal{E}\left[\hat{\mathbf{k}}\times\hat{\mathbf{e}}\right]\left\{a\left(t\right)\exp\left[i\,\vec{\mathbf{k}}\;\;\vec{\mathbf{r}} - i\,\omega\,t\right] - a^{\dagger}\left(t\right)\exp\left[-i\,\vec{\mathbf{k}}\;\;\vec{\mathbf{r}} - i\,\omega\,t\right]\right\} \qquad [\text{ III-3b }]$$

---

[11] In what follows, for simplicity we drop the $\vec{\mathbf{k}}$ subscripts on the operators and state vectors with the obvious meaning that $\left|\left\{n_{\vec{\mathbf{k}}}\right\}\right\rangle \quad |n\rangle$, $a_{\vec{\mathbf{k}}} \quad a$, *etc...*

where $\mathcal{E} = \sqrt{\dfrac{\hbar\omega}{2\varepsilon_0 V}}$

$$\begin{aligned} \langle n|\,\vec{\mathbf{E}}\,|n\rangle &= 0 \\ \langle n|\,\vec{\mathbf{H}}\,|n\rangle &= 0 \end{aligned} \qquad \text{[III-4a]}$$

$$\begin{aligned} \Delta\mathbf{E} &= \sqrt{\left\{\langle n|\,\vec{\mathbf{E}}\;\vec{\mathbf{E}}\,|n\rangle - \langle n|\,\vec{\mathbf{E}}\,|n\rangle^2\right\}} = \sqrt{\frac{\hbar\omega}{\varepsilon_0 V}}\left(n+\tfrac{1}{2}\right)^{1/2} = \sqrt{2}\,\mathcal{E}\left(n+\tfrac{1}{2}\right)^{1/2} \\ \Delta\mathbf{H} &= \sqrt{\left\{\langle n|\,\vec{\mathbf{H}}\;\vec{\mathbf{H}}\,|n\rangle - \langle n|\,\vec{\mathbf{H}}\,|n\rangle^2\right\}} = \sqrt{\frac{\hbar\omega}{\mu_0 V}}\left(n+\tfrac{1}{2}\right)^{1/2} = \sqrt{\frac{\varepsilon_0}{\mu_0}}\,\sqrt{2}\,\mathcal{E}\left(n+\tfrac{1}{2}\right)^{1/2} \qquad \text{[III-4b]} \\ \Delta\mathbf{E}\;\Delta\mathbf{H} &= c\,\frac{\hbar\omega}{V}\left(n+\tfrac{1}{2}\right) = \sqrt{\frac{\varepsilon_0}{\mu_0}}\;2\,\mathcal{E}^2\left(n+\tfrac{1}{2}\right) \end{aligned}$$

c. **Phase of field associated with single mode**:

To obtain something analogous to the classical theory we would like to separate the creation and destruction operators (and, thus, the electric and magnetic field operators) into a product of amplitude and phase operators. Following Susskind and Glogower,[12] we define a ***phase operator***, Φ such that

$$\begin{aligned} a &\equiv \left(\mathcal{N}+1\right)^{1/2}\exp\left(i\,\Phi\right) \\ a^\dagger &\equiv \exp\left(-i\,\Phi\right)\left(\mathcal{N}+1\right)^{1/2} \end{aligned} \qquad \text{[III-5]}$$

Defined in this way, the basic properties of the phase operator may be evaluated from known properties of the creation, destruction and number operators. Inverting, we obtain

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[12] Susskind, L. and Glogower, J., *Physics*, **1**, 49 (1964)

$$\begin{aligned} \exp(i\,\Phi) &\equiv \left(\mathcal{N}+1\right)^{-1/2} a \\ \exp(-i\,\Phi) &\equiv a^{\dagger}\left(\mathcal{N}+1\right)^{-1/2} \end{aligned} \qquad \text{[III-6]}$$

and since $a\,a^{\dagger}=\mathcal{N}+1$, it follows that

$$\exp(i\,\Phi)\,\exp(-i\,\Phi)=1 \qquad \text{[III-7]}$$

but only in this order! Operating on number states with the phase operators, we obtain from Equation [I-26]

$$\begin{aligned} \exp(i\,\Phi)\,|n\rangle &= \left(\mathcal{N}+1\right)^{-1/2} a\,|n\rangle = \left(\mathcal{N}+1\right)^{-1/2}(n)^{1/2}\,|n-1\rangle = |n-1\rangle \\ \exp(-i\,\Phi)\,|n\rangle &= a^{\dagger}\left(\mathcal{N}+1\right)^{-1/2}|n\rangle = a^{\dagger}\left(n+1\right)^{-1/2}|n\rangle = |n+1\rangle \end{aligned} \qquad \text{[III-8]}$$

Consequently, the **only nonvanishing matrix elements** of the phase operator are

$$\begin{aligned} \langle n-1|\exp(i\,\Phi)\,|n\rangle &= 1 \\ \langle n+1|\exp(-i\,\Phi)\,|n\rangle &= 1 \end{aligned} \qquad \text{[III-9]}$$

The phase operators defined by Equation [III-36] do have the felicitous or *classically analogous* property of revealing ***magnitude independent*** information, but unfortunately they are nonHermitian operators -- *i.e.*

$$\langle n-1|\exp(i\,\Phi)\,|n\rangle \neq \langle n|\exp(i\,\Phi)\,|n-1\rangle$$

-- and, hence, **cannot represent observables**. However, they may be **paired** into operators that are observables -- *viz*.

$$\begin{aligned} \cos\Phi &= \frac{1}{2}\left\{ \exp\left(i\,\Phi\right) + \exp\left(-i\,\Phi\right)\right\} \\ \sin\Phi &= \frac{1}{2\,i}\left\{ \exp\left(i\,\Phi\right) - \exp\left(-i\,\Phi\right)\right\} \end{aligned} \qquad \text{[III-10]}$$

which have the following nonvanishing matrix elements:

$$\begin{aligned} \langle n-1|\cos\Phi|n\rangle &= \langle n|\cos\Phi|n-1\rangle = \frac{1}{2} \\ \langle n-1|\sin\Phi|n\rangle &= -\langle n|\sin\Phi|n-1\rangle = \frac{1}{2\,i} \end{aligned} \qquad \text{[III-11]}$$

These *nearly commuting* operators[13] may be adopted as the quantum mechanical operators which represent (as we will demonstrate anon) the observable phase properties of the electromagnetic field.

For the Fock state:

$$\langle n|\cos\Phi|n\rangle = \langle n|\sin\Phi|n\rangle = 0 \qquad \text{[III-12a]}$$

$$\Delta\cos\Phi = \Delta\sin\Phi = \sqrt{\left\{\langle n|\cos^2\Phi|n\rangle - \langle n|\cos\Phi|n\rangle^2\right\}} = \sqrt{1/2} \qquad \text{[III-12b]}$$

$$\Delta\cos\Phi\;\Delta\sin\Phi = 1/2 \qquad \text{[III-12c]}$$

c. **The coordinate or Schrödinger representation of state:**

Recall from Equations [I-10a] and [I-31] that

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[13] Also, it may be easily established that the matrix elements of their commutator are given by

$$\langle n|\left[\cos\Phi,\sin\Phi\right]|n'\rangle = \frac{i}{2}\,\delta_{nn'}\,\delta_{n0}$$

$$\langle q|n\rangle = \frac{1}{\sqrt{n\,!}}\left[\sqrt{\frac{m}{2\,\hbar\omega}}\right]^n \left[\omega\, q - \frac{\hbar}{m}\frac{d}{dq}\right]^n \langle q|0\rangle$$

$$= \sqrt{\frac{1}{2^n\; n!}\sqrt{\frac{\omega}{\pi\hbar}}}\; H_n\left(\sqrt{\frac{m\,\omega}{\hbar}}\; q\right)\exp\left[-\frac{m\,\omega}{2\,\hbar}\, q^2\right] \qquad [\text{ III-13 }]$$

Therefore, the probability $P(q)$ of eigenvalues q for a given Fock state $|n\rangle$ is give by

$$P(q) = |\langle q|\, n\rangle| = \frac{1}{2^n\; n!}\sqrt{\frac{\omega}{\pi\,\hbar}}\; H_n^2\left(\sqrt{\frac{m\,\omega}{\hbar}}\; q\right)\exp\left[-\frac{m\,\omega}{\hbar}\, q^2\right] \qquad [\text{ III-14 }]$$

d. **Approximate "localization" of a photon:** [14]

Of course a plane wave is distributed or "de-localized" in both time and space. Defining the "wave function for a photon" is a task fraught with danger,[15] but the simpler task of defining a wave function approximately localized at a given instant is relatively straight forward -- *viz*.

$$\left|\psi\left(\vec{\mathbf{r}}_0\right)\right\rangle_{\vec{\mathbf{k}}_0\left|\,\Delta\vec{\mathbf{k}}\right|^2} = C\sum_{\vec{\mathbf{k}}}\exp\left[\frac{\left|\vec{\mathbf{k}}-\vec{\mathbf{k}}_0\right|^2}{2\left|\,\Delta\,\vec{\mathbf{k}}\right|^2}\right]\exp\left[i\,\vec{\mathbf{k}}\;\;\vec{\mathbf{r}}_0\right]\left|0,0,0,\ldots n_{\vec{\mathbf{k}}}=1,\ldots,0,0,0\right\rangle \qquad [\text{ III-14 }]$$

2. **Photon States of *Well-defined* Phase:**

Consider the state defined by

$$|\phi\rangle \equiv \lim_{s\to\infty}\left(s+1\right)^{-1/2}\sum_{n=0}^{s}\exp\left[i\,n\;\phi\right]|\,n\rangle \qquad [\text{ III-15 }]$$

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[14] See Section 10.4.2 in Leonard Mandel and Emil Wolf, *Optical Coherence and Quantum Optics*, Cambridge Press (1995), ISBN 0-521-417112.

[15] See Section 1.5.4 in Marlan O. Scully and M. Suhail Zubairy, Quantum Optics, Cambridge Press (1997), ISBN 0-521-43458.

Clearly, $\langle \phi | \phi \rangle = 1$ given the orthonormal properties of the number states. Essential question: Is this state an eigenstate of the phase operators? To answer the question we need to consider the following ***potential eigenvalue equation***:

$$\cos\Phi \,|\phi\rangle = \frac{1}{2} \lim_{s\to\infty} (s+1)^{-1/2} \left\{ \sum_{n=0}^{s} \exp[i\,n\,\phi]\, \exp[i\,\Phi]\, |n\rangle + \sum_{n=0}^{s} \exp[i n\ \phi]\, \exp[-i\ \Phi]\, |n\rangle \right\} \qquad [\text{ III-16a }]$$

Using Equations [III-10] and [III-10], we obtain

$$\begin{aligned} \cos\Phi\,|\phi\rangle &= \frac{1}{2} \lim_{s\to\infty} (s+1)^{-1/2} \left\{ \sum_{n=1}^{s} \exp[i n\,\phi]\, |n-1\rangle + \sum_{n=0}^{s} \exp[i\,n\,\phi]\, |n+1\rangle \right\} \\ &= \frac{1}{2} \lim_{s\to\infty} (s+1)^{-1/2} \left\{ \exp(i\phi) \sum_{\nu=0}^{s-1} \exp[i\,\nu\,\phi]\, |\nu\rangle + \exp[-i\,\phi] \sum_{\nu=1}^{s+1} \exp[i\,\nu\,\phi]\, |\nu\rangle \right\} \qquad [\text{ III-16b }] \\ &= \cos\phi\ |\phi\rangle \\ &\quad + \frac{1}{2} \lim_{s\to\infty} (s+1)^{-1/2} \left\{ \exp[i\,s\,\phi]\, |s+1\rangle - \exp[i\,(s+1)\,\phi]\, |s\rangle - \exp[-i\,\phi]\, |0\rangle \right\} \end{aligned}$$

so that the state $|\phi\rangle$ fails to be a strict eigenket of $\cos\Phi$ by terms that diminish faster than $(s+1)^{-1/2}$ as $s \to \infty$. Similarly, we can see that diagonal matrix elements of $\cos\Phi$ and $\sin\Phi$ are given by

$$\langle \phi | \cos\Phi | \phi \rangle = \cos\phi \left\{ 1 - \lim_{s\to\infty} (s+1)^{-1} \right\} \qquad \cos\phi \qquad [\text{ III-17a }]$$

$$\langle \phi | \sin\Phi | \phi \rangle = \sin\phi \left\{ 1 - \lim_{s\to\infty} (s+1)^{-1} \right\} \qquad \sin\phi \qquad [\text{ III-17b }]$$

Reprise of Characteristics and Properties of Phase States:

a. **The expectation value of the number operator and the *fractional uncertainty* associated with a state of well-defined phase**:

$$\langle\phi|\mathcal{N}|\phi\rangle = \lim_{s\to\infty}(s+1)^{-1}\sum_{n=0}^{s} n = \lim_{s\to\infty}(s+1)^{-1}\left[\frac{s(s+1)}{2}\right] = \lim_{s\to\infty}\frac{s}{2} \qquad \text{[III-18a]}$$

$$\begin{bmatrix}\text{fractional}\\ \text{uncertainty}\end{bmatrix} = \frac{\sqrt{\left\{\langle\phi|\mathcal{N}^{2}|\phi\rangle - \langle\phi|\mathcal{N}|\phi\rangle^{2}\right\}}}{\langle\phi|\mathcal{N}|\phi\rangle}$$

$$= \frac{\sqrt{\left\{\lim_{s\to\infty}(s+1)^{-1}\sum_{n=0}^{s} n^{2} - \left[\lim_{s\to\infty}(s+1)^{-1}\sum_{n=0}^{s} n\right]^{2}\right\}}}{\lim_{s\to\infty}(s+1)^{-1}\sum_{n=0}^{s} n} \qquad \text{[III-18b]}$$

$$= \frac{\sqrt{\left\{\lim_{s\to\infty}\left[\frac{1}{6}\left(2s^{2}+s\right) - \frac{1}{4}s^{2}\right]\right\}}}{\lim_{s\to\infty}\frac{s}{2}} = \frac{1}{\sqrt{3}}$$

b. **Expectation value of the fields associated with a single mode**:

From Equation [III-3a]

$$\langle\phi|\vec{\mathbf{E}}|\phi\rangle = -2\sqrt{\frac{\hbar\,\omega}{2\,\varepsilon_0\,V}}\;\hat{\mathbf{e}}\,\sin\!\left(\vec{\mathbf{k}}\;\;\vec{\mathbf{r}} - \omega\,t + \phi\right)\lim_{s\to\infty}(s+1)^{-1}\sum_{n=0}^{s}(n+1)^{1/2} \qquad \text{[III-19]}$$

diverges as $\sqrt{s}$ for large s !

c. **Phase of field associated with single mode**:

$$\begin{aligned} \langle \phi \,|\, \cos\Phi \,|\phi\rangle &= \cos\phi \\ \langle \phi \,|\, \sin\Phi \,|\phi\rangle &= \sin\phi \end{aligned} \qquad [\text{ III-20a }]$$

$$\Delta\cos\Phi = \Delta\sin\Phi = \sqrt{\left\{\langle \phi \,|\, \cos^2\Phi |\phi\rangle - \langle \phi \,|\cos\Phi\,|\phi\rangle^2\right\}} = 0 \qquad [\text{ III-20b }]$$

d. **Probability of photon number**:

Finally, we may easily deduce the probability of finding n photons (*i.e.* the photon statistics) in a particular state of well defined phase -- *viz.*

$$P_n = \left|\langle n|\phi\rangle\right|^2 \equiv \lim_{s\to\infty}(s+1)^{-1} \qquad [\text{ III-50 }]$$

We see that there is a equal, but small probability of any number: this agrees with the intuition that the magnitude of the field is completely undetermined if the phase is precisely known!

3. ***Coherent*** **Photon States:** [16]

It would, indeed, be useful to have eigenstates of the ***destruction operator*** (electric or magnetic field) -- *viz.*

$$a_{\vec{\mathbf{k}}}\left|\alpha_{\vec{\mathbf{k}}}\right\rangle = \alpha_{\vec{\mathbf{k}}}\left|\alpha_{\vec{\mathbf{k}}}\right\rangle \qquad [\text{ III-51 }]$$

Reprise of Characteristics and Properties of Coherent States:

a. **The Fock state representation of the coherent state**:

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[16] The coherent state is a **Harvard invention**! See R. J. Glauber, Phys. Rev. **131**, 2766 (1963).

Since. $a^{\dagger}|n\rangle = \sqrt{n+1}\,|n+1\rangle$ and $a\;a^{\dagger} = \mathcal{N}+1$, then $\langle n|\,a = \sqrt{n+1}\,\langle n+1|$ and we are able to write a **representative** of the sought state in the number state basis -- *viz*.

$$\langle n|\,a\,|\alpha\rangle = \sqrt{n+1}\,\langle n+1|\alpha\rangle = \alpha\,\langle n|\alpha\rangle \qquad [\text{ III-52a }]$$

or

$$\langle n|\alpha\rangle = \frac{\alpha}{\sqrt{n}}\langle n-1|\alpha\rangle = \frac{\alpha^{n}}{\sqrt{n!}}\langle 0|\alpha\rangle \qquad [\text{ III-52b }]$$

Using the expansion of the identity operator, the eigenket becomes

$$|\alpha\rangle = \sum_{n}|n\rangle\langle n|\alpha\rangle = \langle 0|\alpha\rangle\;\sum_{n}\frac{\alpha^{n}}{\sqrt{n!}}\,|n\rangle\ . \qquad [\text{ III-53 }]$$

To normalize the eigenket write

$$\langle\alpha|\alpha\rangle = \langle\alpha|0\rangle\langle 0|\alpha\rangle\sum_{n}\frac{\alpha^{\;n}\;\alpha^{n}}{n!} = \langle\alpha|0\rangle\langle 0|\alpha\rangle\;\exp\left[|\alpha|^{2}\right] = 1 \qquad [\text{ III-54 }]$$

so that $\langle\alpha|0\rangle = \langle 0|\alpha\rangle = \exp\left[\left(-\frac{1}{2}|\alpha|^{2}\right)\right]$. Finally, we see that

$$|\alpha\rangle = \exp\left[-\frac{1}{2}|\alpha|^{2}\right]\sum_{n}\frac{\alpha^{n}}{\sqrt{n!}}\,|n\rangle \qquad [\text{ III-55 }]$$

is a normalize representation of the eigenkets of the destruction operator.

b. **The expectation value of the number operator and the *fractional uncertainty* associated with a coherent state**:

$$\langle \alpha | \mathcal{N} | \alpha \rangle = |\alpha|^2 \qquad \text{[III-56a]}$$

$$\begin{aligned}\begin{bmatrix}\text{fractional}\\ \text{uncertainty}\end{bmatrix} &= \frac{\sqrt{\left\{\langle \alpha | \mathcal{N}^{2} | \alpha\rangle - \langle \alpha | \mathcal{N} | \alpha\rangle^{2}\right\}}}{\langle \alpha | \mathcal{N} | \alpha\rangle} = \frac{1}{|\alpha|^2}\sqrt{\left\{\exp\left(-|\alpha|^2\right)\sum \frac{|\alpha|^{2n}}{n!}\, n^2 - |\alpha|^4\right\}} \\ &= \frac{1}{|\alpha|^2}\sqrt{\left\{\exp\left(-|\alpha|^2\right)\sum \frac{|\alpha|^{2n}}{n!}\left[n\left(n-1\right)+n\right] - |\alpha|^4\right\}} \\ &= |\alpha|^{-1}\end{aligned} \qquad \text{[III-56b]}$$

Thus, we see that the fractional uncertainty diminishes with mean photon number!

c. **Expectation value of the electric field associated with a single mode**:

From Equation [III-3a]

$$\langle \alpha | \vec{\mathbf{E}} | \alpha \rangle = -2\sqrt{\frac{\hbar\,\omega}{2\,\varepsilon_0\,V}}\;\hat{\mathbf{e}}\,|\alpha|\,\sin\left(\vec{\mathbf{k}}\cdot\vec{\mathbf{r}} - \omega\,t + \vartheta\right) \qquad \text{[III-57a]}$$

where $\alpha = |\alpha|\,\exp\left(i\,\vartheta\right)$.

$$\Delta\mathbf{E} = \sqrt{\left\{\langle \alpha | \vec{\mathbf{E}}\cdot\vec{\mathbf{E}} | \alpha\rangle - \langle \alpha | \vec{\mathbf{E}} | \alpha\rangle^{2}\right\}} = \sqrt{\frac{\hbar\,\omega}{2\,\varepsilon_0\,V}}\,[17] \qquad \text{[III-57b]}$$

---

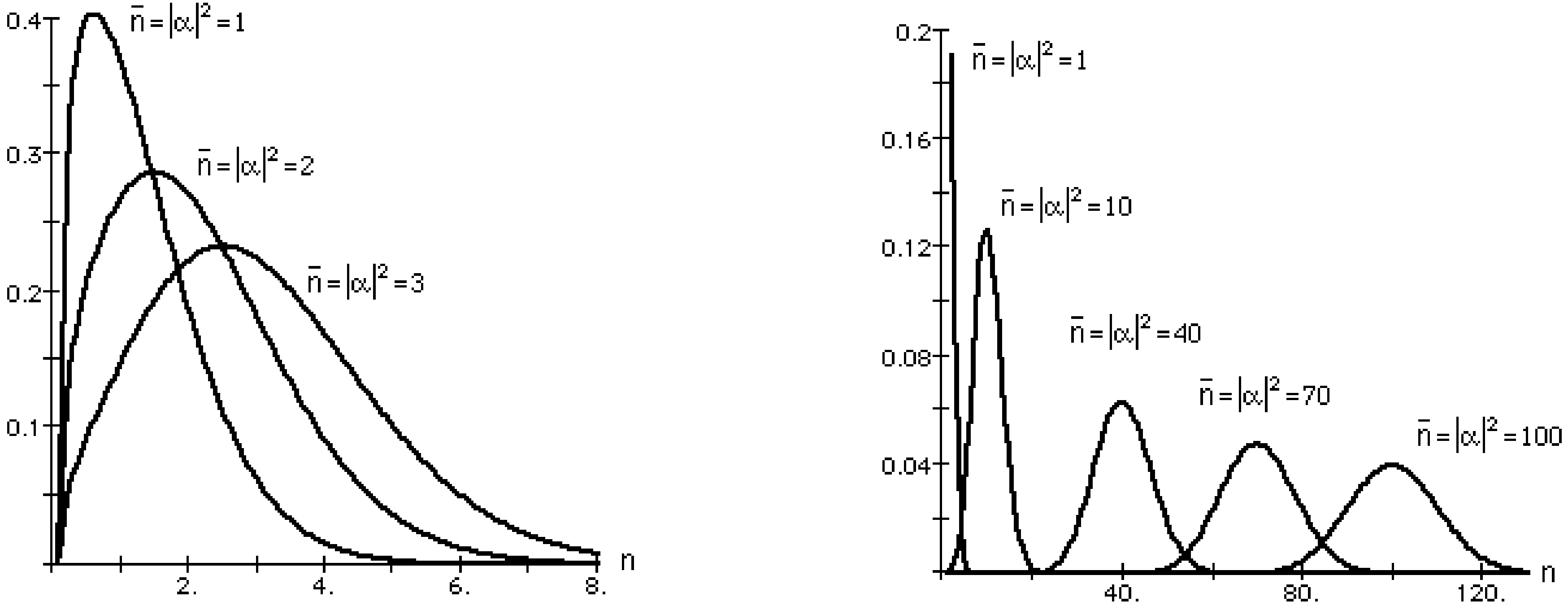
[17] Similarly $\Delta\mathbf{H} = \dfrac{1}{c\,\mu_0}\sqrt{\dfrac{\hbar\,\omega}{2\,\varepsilon_0\,V}}$ for the coherent state, so that $\Delta\mathbf{E}\;\Delta\mathbf{H} = c\,\hbar\,\omega/2\,V$.

d. **Probability of photon number**:

From the representation of the coherent state given in Equation [III-55] we may easily deduce the probability of finding n photons (the photon statistics) in a particular coherent state is given by a **Poisson distribution** characterized by the mean value $\bar{n} = |\alpha|^2$. -- *viz*.

$$P_n = \left|\langle n|\alpha\rangle\right|^2 = \exp\left[-|\alpha|^2\right] \frac{|\alpha|^{2n}}{n!} \qquad \text{[III-58]}$$

SAMPLE POISSON DISTRIBUTIONS - COHERENT STATE PHOTON STATISTICS



e. **Phase of field associated with single mode**:

$$
\begin{aligned}
\langle \alpha | \cos\Phi | \alpha \rangle &= \frac{1}{2}\exp\left[-\frac{1}{2}|\alpha|^2\right]\sum_{n'}\sum_{n}\langle n'|\frac{\alpha^{n'}}{\sqrt{n'!}}\left[(\mathcal{N}+1)^{-1/2}\,a + a^{\dagger}\,(\mathcal{N}+1)^{-1/2}\right]\frac{\alpha^{n}}{\sqrt{n!}}|n\rangle \\
&= \frac{1}{2}\exp\left[-\frac{1}{2}|\alpha|^2\right]\sum_{n}\left[\frac{\alpha^{n+1}\,\alpha^{n} + \alpha^{n}\,\alpha^{n+1}}{\sqrt{\{(n+1)!\,n!\}}}\right] \\
&= |\alpha|\cos\vartheta\,\exp\left[-\frac{1}{2}|\alpha|^2\right]\sum_{n}\left[\frac{|\alpha|^{2n}}{n!\,\sqrt{(n+1)}}\right]
\end{aligned}
\qquad \text{[III-59a]}
$$

Unfortunately, it is not possible to evaluate this summation analytically. However, Carruthers[18] has given an asymptotic expansion which is valid for a large mean number of photons -- *viz.*

$$
\langle \alpha | \cos\Phi | \alpha \rangle = \cos\vartheta\,\left[1 - \frac{1}{8|\alpha|^2} + \ldots\right] \qquad |\alpha|^2 >> 1 \qquad \text{[III-59b]}
$$

f. **Coherent states as a basis:**

As we will see presently, the coherent states are very useful in describing the quantized electromagnetic field, but, alas, there is a complication -- **the coherent states are not truly orthogonal!** From Equation [III-6] we see that

$$
\begin{aligned}
\langle \beta | \alpha \rangle &= \exp\left[-\frac{1}{2}|\alpha|^2 - \frac{1}{2}|\beta|^2\right]\sum_{n}\frac{\beta^{n}\alpha^{n}}{n!} \\
&= \exp\left[-\frac{1}{2}|\alpha|^2 - \frac{1}{2}|\beta|^2 + \alpha\,\beta\right]
\end{aligned}
\qquad \text{[III-60]}
$$

so that

---

[18] Carruthers, P. and Nieto, M. M., *Phys. Rev. Lett.* **14**, 387 (1965)

$$\begin{aligned}\langle\alpha|\beta\rangle\langle\beta|\alpha\rangle &= \exp\left(-|\alpha|^2-|\beta|^2+\alpha\beta\ \ +\alpha\ \ \beta\right)\\ &= \exp\left(-(\alpha-\beta)(\alpha\ \ -\beta\ \ )\right)=\exp\left(-|\alpha-\beta|^2\right)\end{aligned} \qquad [\ \text{III-61}\ ]$$

That is, t**he eigenkets are approximately orthogonal** only when $|\alpha-\beta|$ is large!

g. **The “displacement operator:”**

There are a growing and significant set of applications where it is useful to express the coherent states directly in terms of the vacuum state $|0\rangle$. If we use the number state generating rule

$$|n\rangle=\frac{\left(a^{\dagger}\right)^{n}}{\sqrt{n!}}|0\rangle$$

-- *i.e.* Equation [I-27] -- the coherent state may be written in the form

$$|\alpha\rangle=\exp\left(-\frac{1}{2}|\alpha|^2\right)\sum_n\frac{\left(\alpha\, a^{\dagger}\right)^{n}}{n!}|0\rangle=\exp\left(\alpha\, a^{\dagger}-\frac{1}{2}|\alpha|^2\right)|0\rangle \qquad [\ \text{III-62}\ ]$$

If we make us of the Baker-Hausdorff theorem,[19] we may easily show that

---

[19] The Baker-Hausdorff theorem or identity may be stated as

$$\exp\{\mathcal{A}+\mathcal{B}\}=\exp\{\mathcal{A}\}\ \exp\{\mathcal{B}\}\ \exp\{-\tfrac{1}{2}[\mathcal{A},\mathcal{B}]\}$$

when $\left[\mathcal{A},[\mathcal{A},\mathcal{B}]\right]=\left[\mathcal{B},[\mathcal{A},\mathcal{B}]\right]=0$. For a proof, see, for example, Charles P. Slichter’s *Principles of Magnetic Resonance*, Appendix A or William Louisell’s *Radiation and Noise in Quantum Electronics*.

$$|\alpha\rangle = \mathcal{A}^{\dagger}(\alpha)\,|0\rangle = \exp\left(\alpha\, a^{\dagger} - \alpha\;\; a\right)|0\rangle \qquad [\text{ III-63 }]$$

so that $\mathcal{A}^{\dagger}(\alpha)$ may be interpreted as a *creation* operator which generates a coherent state from the vacuum. (Its adjoint operator $\mathcal{A}(\alpha) = \mathcal{A}^{\dagger}(-\alpha)$ is a *destruction* operator which destroys a state). In some treatments $\mathcal{A}^{\dagger}(\alpha)$ is described as the "displacement operator" (written $\mathcal{D}(\alpha)$)[20] and the coherent states are called the "displaced states of the vacuum." [21]

To explore this point of view (and to give some meaning to the phase of the coherent state eigenvalue), we may express $|\alpha\rangle$ in a two-dimensional, dimensionless "phase space" representation. To that end, following Equation [I-16], we write the dimensionless coordinate as

$$\theta = \left(\frac{m\omega}{\hbar}\right)^{1/2} q = a^{\dagger}\exp[i\,\gamma] + a\;\exp[-i\,\gamma] \qquad [\text{ III-64a }]$$

and the dimensionless momentum as

$$\pi = \left(\frac{2}{m\,\hbar\,\omega}\right)^{1/2} p = a^{\dagger}\exp\left[i\left(\gamma + \pi/2\right)\right] + a\;\exp\left[-i\left(\gamma + \pi/2\right)\right] \qquad [\text{ III-64b }]$$

so that

$$[\theta, \pi] = 2\,i\left[a, a^{\dagger}\right] = 2\,i \qquad [\text{ III-64c }]$$

---

[20] We can (or rather you will) show that $\mathcal{D}^{\dagger}(\alpha)\, a\; \mathcal{D}(\alpha) = a + \alpha$ and $\mathcal{D}^{\dagger}(\alpha)\, a^{\dagger}\, \mathcal{D}(\alpha) = a^{\dagger} + \alpha$

[21] See *Elements of Quantum Optics*, Pierre Meystre and Murray Sargent III, Spinger-Verlag (1991), ISBN 0-387-54190-X.

and since these variables are canonical [22]

$$\left\langle (\Delta\theta)^2 \right\rangle \left\langle (\Delta\pi)^2 \right\rangle \geq 1 \qquad \text{[III-64d]}$$

Since

$$\begin{aligned} a^{\dagger} &= \frac{1}{2}\left(\theta - i\,\pi\right)\exp\left[-i\,\gamma\right] \\ a &= \frac{1}{2}\left(\theta + i\,\pi\right)\exp\left[i\,\gamma\right] \end{aligned} \qquad \text{[III-65]}$$

the mode field (see Equation [II-24a]) b

$$\vec{\mathbf{E}}\left(\vec{\mathbf{r}},t\right) = i\,\hat{\mathbf{e}}\,\mathcal{E}\left\{ a\;\exp\left[i\,\vec{\mathbf{k}}\cdot\vec{\mathbf{r}} - i\,\omega\,t\right] - a^{\dagger}\;\exp\left[-i\,\vec{\mathbf{k}}\cdot\vec{\mathbf{r}} + i\,\omega\,t\right]\right\} \qquad \text{[III-66a]}$$

becomes

$$\vec{\mathbf{E}}\left(\vec{\mathbf{r}},t\right) = -\,\hat{\mathbf{e}}\,\mathcal{E}\left\{\pi\,\cos\left(\vec{\mathbf{k}}\cdot\vec{\mathbf{r}} - \omega\,t + \gamma\right) + \theta\,\sin\left(\vec{\mathbf{k}}\cdot\vec{\mathbf{r}} - \omega\,t + \gamma\right)\right\} \qquad \text{[III-66b]}$$

Since p has a coordinate space representation $-i\,\hbar\,d/dq = -i\left(\hbar\,\omega/2\right)^{1/2} d/d\theta$ and q has a momentum representation $i\,\hbar\,d/dp = i\left(\hbar/2\,\omega\right)^{1/2} d/d\pi$, [23]

$$\begin{aligned} \alpha\,a^{\dagger} - \alpha\;a &= \alpha_r\left(a^{\dagger} - a\right) + i\,\alpha_i\left(a^{\dagger} + a\right) \\ &= -\left[\alpha_r\;d/d\theta + \alpha_i\;d/d\pi\right] \end{aligned} \qquad \text{[III-67a]}$$

---

[22] Of course, in general $\left\langle (\Delta\mathbf{A})^2 \right\rangle \left\langle (\Delta\mathbf{B})^2 \right\rangle \geq \frac{1}{2}\left|\left\langle \left[\,\mathbf{A},\mathbf{B}\,\right] \right\rangle\right|^2$ where $\left\langle (\Delta\mathbf{A})^2 \right\rangle = \left\langle \mathbf{A}^2 \right\rangle - \left\langle \mathbf{A} \right\rangle^2$

[23] If this unfamiliar, see Equations [I-20] and [I-22] in the lecture notes entitled *The Interaction of Radiation and Matter: Semiclassical Theory*.

and

$$\mathcal{A}^{\dagger}(\alpha) = \exp\left(\alpha\, a^{\dagger} - \alpha\ \ a\right) = \exp\left[-\left(\alpha_r\ d/d\theta + \alpha_i\ d/d\pi\right)\right] \qquad \text{[III-67b]}$$

Thus, $\mathcal{A}^{\dagger}(\alpha)$ defines or generates a two-dimensional Taylor expansion when it acts on a function of θ and π. In particular, if we take the "phase space" representation of the ground or vacuum state $|\theta\pi\rangle$ as the product of two Gaussians (see Equations [I-10a] and [I-29]), then $\mathcal{A}^{\dagger}(\alpha)|\theta\pi\rangle$ represents a shift or displacement of this "phase space" representation -- *i.e.*

$$\langle\theta\pi|\alpha\rangle = \langle\theta\pi|\mathcal{A}^{\dagger}(\alpha)|0\rangle = u_G(\theta - \alpha_r)\ u_G(\pi - \alpha_i) \qquad \text{[III-68]}$$

In light of Equation [II-23b], $|\alpha(t)\rangle = |\alpha \exp(-i\,\omega\, t)\rangle$ we can write

$$\langle\theta\pi|\alpha(t)\rangle = u_G\big(\theta - |\alpha|\cos(\omega\, t + \varphi)\big)\ u_G\big(\pi - |\alpha|\cos(\omega\, t + \varphi)\big) \qquad \text{[III-69]}$$

where $\alpha = |\,\alpha\,|\exp(i\,\varphi)$.

h. The diagonal coherent-state representation of the density operator (Glauber-Sudarshan P-representation):

It may be easily established that

$$\overline{\overline{\mathbf{1}}} = \frac{1}{\pi}\iint|\beta\rangle\langle\beta|\,d^2\beta = \iint|\beta\rangle\langle\beta|\,d\,\mathbf{Re}(\beta)\,d\,\mathbf{Im}(\beta) \qquad \text{[III-70]}$$

so that it seems quite reseasonable to look for a representation of the density matrix is the form

$$\overline{\overline{\rho}} = \iint P(\,\beta\,)\,|\beta\rangle\langle\beta|\,d^2\beta \qquad \text{[III-71]}$$

For a pure coherent state, P is clearly a two-dimensional delta function

Example 1 -- Coherent state

$$P(\beta) = \delta^{(2)}(\beta - \alpha) = \delta^{(1)}\left(\mathbf{Re}(\beta) - \mathbf{Re}(\alpha)\right)\delta^{(1)}\left(\mathbf{Im}(\beta) - \mathbf{Im}(\alpha)\right) \qquad [\text{ III-72 }]$$

In general, using Equation [III-60] -- *i.e.*

$$\langle\beta|\alpha\rangle = \exp\left[-\frac{1}{2}|\alpha|^2 - \frac{1}{2}|\beta|^2 + \alpha\beta\right] \qquad [\text{ III-60 }]$$

we may find a simple procedure for finding the P-representation by writing

$$\begin{aligned}\langle -\alpha|\,\overline{\rho}\,|\alpha\rangle &= \iint P(\beta)\langle -\alpha|\beta\rangle\langle\beta|\alpha\rangle\, d^2\beta \\ &= \exp\left(-|\alpha|^2\right)\iint\left[P(\beta)\exp\left(-|\beta|^2\right)\right]\exp\left[\alpha\beta - \beta\alpha\right] d^2\beta\end{aligned} \qquad [\text{III-73 }]$$

Thus, $\langle -\alpha|\,\overline{\rho}\,|\alpha\rangle\exp\left(-|\alpha|^2\right)$ is the two-dimensional Fourier transform 0f the function $P(\beta)\exp\left(-|\beta|^2\right)$ and we may write

$$P(\beta) = \frac{1}{\pi^2}\exp\left(|\beta|^2\right)\iint\langle -\alpha|\,\overline{\rho}\,|\alpha\rangle\exp\left(|\alpha|^2\right)\exp\left[-\alpha\beta + \beta\alpha\right] d^2\alpha \qquad [\text{ III-74 }]$$

As a second example, consider a thermal radiation field described by a canonical ensemble

$$\overline{\rho} = \frac{\exp\left(-\mathcal{H}/k_B T\right)}{\mathrm{Tr}\left[\exp\left(-\mathcal{H}/k_B T\right)\right]} \qquad [\text{III-75 }]$$

where $\mathcal{H} = \hbar\omega\left\{a^\dagger a + \frac{1}{2}\right\}$. Thus,

$$\overline{\rho} = \sum_n\left[1 - \exp\left(-\frac{\hbar\omega}{k_B T}\right)\right]\exp\left(-\frac{n\,\hbar\omega}{k_B T}\right)|n\rangle\langle n| \qquad [\text{III-76 }]$$

and $$\langle n\rangle = \mathrm{Tr}\left[\overline{\rho}\, a^{\dagger} a\right] = \sum_n \left[\exp\left(\frac{\hbar\omega}{k_B T}\right) - 1\right]^{-1} \qquad \text{[III-77]}$$

so that $$\overline{\rho} = \sum_n \frac{\langle n\rangle^n}{\left(1+\langle n\rangle\right)^{n+1}} |n\rangle\langle n| \qquad \text{[III-78]}$$

Thus, we can write $$\langle n|\,\overline{\rho}\,|n\rangle = \frac{\langle n\rangle^n}{\left(1+\langle n\rangle\right)^{n+1}} \qquad \text{[III-79]}$$

and

$$\begin{aligned} \langle -\alpha|\,\overline{\rho}\,|\alpha\rangle &= \sum_n \frac{\langle n\rangle^n}{\left(1+\langle n\rangle\right)^{n+1}} \langle -\alpha|n\rangle\langle n|\alpha\rangle \\ &= \frac{\exp\left(-|\alpha|^2\right)}{1+\langle n\rangle} \sum_n \frac{\left(-|\alpha|^2\right)^n}{n!} \left[\frac{\langle n\rangle}{\left(1+\langle n\rangle\right)}\right]^n \\ &= \frac{\exp\left(-|\alpha|^2\right)}{1+\langle n\rangle} \exp\left[-|\alpha|^2 \Big/ \left(1+\frac{1}{\langle n\rangle}\right)\right] \end{aligned} \qquad \text{[III-80]}$$

Finally, we see that

Example 2 -- Thermal radiation - a chaotic state

$$\begin{aligned} P(\alpha) &= \frac{\exp\left(|\alpha|^2\right)}{\pi^2\left(1+\langle n\rangle\right)} \iint \exp\left[-|\beta|^2 \Big/ \left(1+\frac{1}{\langle n\rangle}\right)\right] \exp\left(-\beta\alpha^* + \alpha\beta^*\right) d^2\beta \\ &= \frac{1}{\pi\langle n\rangle} \exp\left(-|\alpha|^2/\langle n\rangle\right) \end{aligned} \qquad \text{[III-81]}$$

As a third example, consider Fock or number state. From Equation [III-55] we see that

$$\langle -\alpha |\, \bar{\rho}\, | \alpha \rangle = \langle -\alpha | n \rangle \langle n | \alpha \rangle = \frac{\exp\left(-|\alpha|^2\right)}{n\,!} \left(-|\alpha|^2\right)^n \qquad \text{[III-82a]}$$

and

$$\begin{aligned} P(\beta) &= \frac{1}{n\,!}\frac{1}{\pi^2} \exp\left(|\beta|^2\right) \iint \left(-|\alpha|^2\right)^n \exp\left[-\alpha\beta \ +\beta\alpha\ \right] d^2\alpha \\ &= \frac{\exp\left(|\beta|^2\right)}{n\,!} \frac{\partial^{\,2n}}{\partial\beta^{*n}\,\partial\beta^{\;n}} \frac{1}{\pi^2} \iint \exp\left[-\alpha\beta \ +\beta\alpha\ \right] d^2\alpha \end{aligned} \qquad \text{[III-82b]}$$

so that

Example 3 -- Pure Fock or number state

$$P(\beta) = \frac{\exp\left(|\beta|^2\right)}{n!} \frac{\partial^{\,2n}}{\partial\beta^{*n}\,\partial\beta^{\;n}} \delta^{(2)}(\beta) \qquad \text{[III-82b]}$$

i. The Glauber-Sudarshan-Klauder “optical equivalence” theorem:

Suppose we have some “normally ordered” function

$$f^{(N)}\left(a, a^{\dagger}\right) = \sum_n \sum_m c_{nm}\; a^{\dagger n}\, a^{m} \qquad \text{[III-83]}$$

The expectation value is given by

$$\left\langle f^{(N)}\left(a, a^{\dagger}\right)\right\rangle = \mathrm{Tr}\left[\bar{\rho}\; f^{(N)}\left(a, a^{\dagger}\right)\right] \qquad \text{[III-84]}$$

Using Equation [III-71] we see that

$$\left\langle f^{(N)}\left(a, a^{\dagger}\right)\right\rangle = \mathrm{Tr}\left[\iint P(\alpha) \sum_{n}\sum_{m} c_{nm} \,|\alpha\rangle\langle\alpha|\, a^{\dagger n} a^{m}\, d^{2}\alpha\right]$$

$$= \iint P(\alpha) \sum_{n}\sum_{m} c_{nm} \langle\alpha|\, a^{\dagger n} a^{m} \,|\alpha\rangle\, d^{2}\alpha \qquad \text{[III-85a]}$$

$$= \iint P(\alpha) \sum_{n}\sum_{m} c_{nm}\, \alpha^{*n} \alpha^{m}\, d^{2}\alpha$$

or, finally, the **“optical equivalence” theorem**

$$\left\langle f^{(N)}\left(a, a^{\dagger}\right)\right\rangle = \iint P(\alpha)\, f^{(N)}\left(\alpha, \alpha^{*}\right) \qquad \text{[III-85b]}$$

j. The Uncertainty Relationship for $\{\theta, \pi\}$:

Since $\left[a, a^{\dagger}\right]=1$ we see from Equation [III-64a] that

$$\left\langle\Delta\theta^{2}\right\rangle = \left\langle\theta^{2}\right\rangle - \left\langle\theta\right\rangle^{2}$$

$$= \left\langle a^{\dagger} a^{\dagger}\right\rangle \exp[2 i \gamma] + \left\langle a\, a\right\rangle \exp[-2 i \gamma] + \left\langle a^{\dagger} a\right\rangle + \left\langle a\, a^{\dagger}\right\rangle$$

$$- \left\langle a^{\dagger}\right\rangle^{2} \exp[2 i \gamma] - \left\langle a\right\rangle^{2} \exp[2 i \gamma] - 2\left\langle a^{\dagger}\right\rangle\left\langle a\right\rangle \qquad \text{[III-86]}$$

$$= \left\langle :\Delta\theta^{2}: \right\rangle + 1$$

where $\langle : \mathbf{A} : \rangle$ symbollizes the normally ordered expectation value of the operator $\mathbf{A}$. From Equation [III-85b]

$$\left\langle :\Delta\theta^{2}\left(a, a^{\dagger}\right): \right\rangle = \iint P(\alpha)\, \Delta\theta^{2}\left(\alpha, \alpha^{*}\right) d^{2}\alpha \qquad \text{[III-87]}$$

$$\left\langle :\Delta\theta^{2}\left(a, a^{\dagger}\right): \right\rangle = \iint P(\alpha)\left[\Delta\alpha^{*}\exp(i\gamma) + \Delta\alpha\exp(-i\gamma)\right]^{2} d^{2}\alpha \qquad [\text{ III-88 }]$$

If we choose γ (and $P(\alpha)$) such that $\left\langle :\Delta\theta^{2}\left(a, a^{\dagger}\right): \right\rangle < 0$, then $\left\langle \Delta\theta^{2} \right\rangle > 1$ and $\left\langle \Delta\pi^{2} \right\rangle > 1$(**squeezed states**)!

IV. INTERACTION HAMILTONIAN -- COUPLING FIELDS AND CHARGES:[23]

To build a complete quantum picture of the interaction of matter and radiation our first and most critical task is to construct a *reliable* Lagrangian-Hamiltonian formulation of the problem. In this treatment, we will confine ourselves to a **nonrelativistic view** which, fortunately, is adequate for most circumstances. We start with a representation of the Lorentz force for a single charged particle -- *viz*.

$$\vec{\mathbf{f}} = q\left\{ \vec{\mathbf{E}} + \vec{\mathbf{v}} \times \vec{\mathbf{B}} \right\} \qquad [\text{ IV-1a }]$$

or in terms of the electromagnetic potentials

$$\vec{\mathbf{f}} = q\left\{ -\vec{\nabla}\phi - \frac{\partial \vec{\mathbf{A}}}{\partial t} + \vec{\mathbf{v}} \times \left(\vec{\nabla} \times \vec{\mathbf{A}} \right) \right\} \qquad [\text{ IV-1b }]$$

Let us now write the **total time derivative** of a component of the vector potential

$$\frac{d\,\mathrm{A}_\alpha}{dt} = \frac{\partial\,\mathrm{A}_\alpha}{\partial\,\mathrm{r}_\beta}\frac{d\,\mathrm{r}_\beta}{dt} + \frac{\partial\,\mathrm{A}_\alpha}{\partial\,t} = \left(\vec{\mathbf{v}} \cdot \vec{\nabla} \right) \mathrm{A}_\alpha + \frac{\partial\,\mathrm{A}_\alpha}{\partial\,t} \qquad [\text{ IV-2 }]$$

so that

$$\vec{\mathbf{v}} \times \left(\vec{\nabla} \times \vec{\mathbf{A}} \right) = \vec{\nabla}\left(\vec{\mathbf{v}} \cdot \vec{\mathbf{A}} \right) - \left(\vec{\mathbf{v}} \cdot \vec{\nabla} \right) \vec{\mathbf{A}} = \vec{\nabla}\left(\vec{\mathbf{v}} \cdot \vec{\mathbf{A}} \right) - \frac{d\,\vec{\mathbf{A}}}{dt} + \frac{\partial\,\vec{\mathbf{A}}}{\partial\,t} \qquad [\text{ IV-3 }]$$

Therefore, we may write the Lorentz force as

$$\mathrm{f}_\alpha = q\left\{ -\frac{\partial}{\partial \mathrm{r}_\alpha}\left[\phi - \mathrm{v}_\beta\,\mathrm{A}_\beta \right] - \frac{d\,\mathrm{A}_\alpha}{dt} \right\} \qquad [\text{ IV-4 }]$$

---

[23] Much of what follows draws heavily on material in Chapter 5 of Rodney Loudon's *The Quantum Theory of Light* (second edition), Oxford (1983). Marlan O. Scully and M. Suhail Zubairy in Section 5.1 of *Quantum Optics*, Cambridge (1997) have a slightly different, but quite insightful treatment of the subject of the atom-field interaction Hamiltonian.

We may also write $$-\mathrm{A}_\alpha = \frac{\partial}{\partial\,\mathrm{v}_\alpha}\left[\phi - \mathrm{v}_\beta\ \mathrm{A}_\beta\right]$$

so that Equation [IV-4] becomes

$$\begin{aligned} \mathrm{f}_\alpha &= -\frac{\partial}{\partial \mathrm{r}_\alpha}\left\{ q\left[\phi - \mathrm{v}_\beta\ \mathrm{A}_\beta\right]\right\} + \frac{d}{dt}\frac{\partial}{\partial \mathrm{v}_\alpha}\left\{ q\left[\phi - \mathrm{v}_\beta\ \mathrm{A}_\beta\right]\right\} \\ &= m\,\frac{d}{dt}\mathrm{v}_\alpha = m\,\frac{d}{dt}\frac{\partial}{\partial \mathrm{v}_\alpha}\left[\frac{1}{2}\mathrm{v}_\beta\ \mathrm{v}_\beta\right] = \frac{d}{dt}\frac{\partial}{\partial \mathrm{v}_\alpha}\left[\frac{1}{2}m\mathrm{v}^2\right] \end{aligned} \qquad [\ \text{IV-5}\ ]$$

which may, in turn, be written

$$\frac{d}{dt}\left\{\frac{\partial}{\partial \mathrm{v}_\alpha}\left[\frac{1}{2}m\mathrm{v}^2 - q\left(\phi - \mathrm{v}_\beta\ \mathrm{A}_\beta\right)\right]\right\} - \frac{\partial}{\partial \mathrm{r}_\alpha}\left[\frac{1}{2}m\mathrm{v}^2 - q\left(\phi - \mathrm{v}_\beta\ \mathrm{A}_\beta\right)\right] = 0\ . \qquad [\ \text{IV-6}\ ]$$

This last equation may now be compared to the **Lagrangian equation of motion** -- *i.e.*

$$\frac{d}{dt}\left\{\frac{\partial}{\partial \mathrm{v}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right)\right\} - \frac{\partial}{\partial \mathrm{r}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) = 0 \qquad [\ \text{IV-7a}\ ]$$

where, in general,

$$\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) = \left[\text{Kinetic Energy}\right] - \left[\text{Potential Energy}\right] = \mathcal{T}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) - \mathcal{U}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right)\ . \qquad [\ \text{IV-7b}\ ]$$

Therefore, we identify

$$\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) = \left[\frac{1}{2}m\mathrm{v}^2 - q\left(\phi - \vec{\mathbf{v}}\ \vec{\mathbf{A}}\right)\right] \qquad [\ \text{IV-8}\ ]$$

as the Lagrangian for a single charged particle. We may write

$$\frac{d}{dt}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) = \frac{\partial}{\partial \mathrm{r}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right)\mathrm{v}_\alpha + \frac{\partial}{\partial \mathrm{v}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right)\dot{\mathrm{v}}_\alpha \qquad [\ \text{IV-9a}\ ]$$

which, in light of Equation [IV-8], becomes

$$\begin{aligned}\frac{d}{dt}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) &= \frac{d}{dt}\left\{\frac{\partial}{\partial \mathrm{v}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right)\right\}\mathrm{v}_\alpha + \frac{\partial}{\partial \mathrm{v}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right)\dot{\mathrm{v}}_\alpha \\ &= \frac{d}{dt}\left\{\mathrm{v}_\alpha\frac{\partial}{\partial \mathrm{v}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right)\right\}\end{aligned} \quad . \qquad \text{[IV-9b]}$$

Therefore,

$$\mathrm{v}_\alpha\frac{\partial}{\partial \mathrm{v}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) - \mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) = \textbf{A constant of motion} \qquad \text{[IV-9c]}$$

and, using the cannonically conjugate momenta associated Equation [IV-8] -- *i.e.*

$$\mathcal{P}_\alpha = \frac{\partial}{\partial \mathrm{v}_\alpha}\mathcal{L}\left(\vec{\mathbf{r}},\vec{\mathbf{v}}\right) = m\mathrm{v}_\alpha + q\mathrm{A}_\alpha \text{ --,} \qquad \text{[IV-10]}$$

we see that Equation [IV-9c] can be written

$$\begin{aligned}\frac{1}{m}\left[\mathcal{P}_\alpha - q\mathrm{A}_\alpha\right]\mathcal{P}_\alpha - \frac{1}{2m}\left[\mathcal{P}_\alpha - q\mathrm{A}_\alpha\right]\left[\mathcal{P}_\alpha - q\mathrm{A}_\alpha\right] + q\phi - \frac{q}{m}\mathrm{A}_\alpha\left[\mathcal{P}_\alpha - q\mathrm{A}_\alpha\right] \\ = \textbf{A constant of motion}\end{aligned} \qquad \text{[IV-11a]}$$

or finally

$$q\phi + \frac{1}{2m}\left[\vec{\mathcal{P}} - q\vec{\mathbf{A}}\right]^2 = \textbf{A constant of motion} \quad . \qquad \text{[IV-11b]}$$

We are now in a position to set forth the **nonrelativistic Hamiltonian of a single charged particle** -- *viz.*

$$\mathcal{H} = q\phi + \frac{1}{2m}\left[\vec{\mathcal{P}} - q\vec{\mathbf{A}}\right]^2 \qquad \text{[IV-12]}$$

with the canonical conjugate variables given by

$$\dot{r}_\alpha = \frac{\partial \mathcal{H}}{\partial \mathcal{P}_\alpha} = \frac{1}{m}\left[\mathcal{P}_\alpha - q\mathrm{A}_\alpha\right] = \mathrm{v}_\alpha \qquad \text{[IV-13a]}$$

$$\dot{\mathcal{P}}_\alpha = -\frac{\partial \mathcal{H}}{\partial r_\alpha} = -q\frac{\partial \phi}{\partial r_\alpha} + \frac{1}{m}\left[\mathcal{P}_\beta - q\mathrm{A}_\beta\right]\frac{\partial}{\partial r_\alpha}\left[q\mathrm{A}_\beta\right] \qquad \text{[IV-13b]}$$

In principal, we are **done**, since we may now write the complete Hamiltonian for a many particle material system as

$$\mathcal{H} = \sum_i \mathcal{H}_i = \sum_i \left\{ q_i\,\phi + \frac{1}{2m_i}\left[\vec{\mathcal{P}}_i - q\,\vec{\mathbf{A}}\right]^2 \right\} \qquad \text{[IV-14]}$$

Unfortunately, this form of the Hamiltonian is not the most useful in optical physics since it is expressed in terms of the vector potential and needs to be evaluated at all charge positions. Most annoyingly, it does not yield an interaction term in the form used earlier -- *i.e.* Equation [III-3a] in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*.

THE MULTIPOLE EXPANSION OF THE CLASSICAL HAMILTONIAN:

Before we manipulate the Hamiltonian further, we digress a bit to get a better fix on what we are really look for. From the most basic notions of electrostatics, the energy of interaction **with an external transverse field** should be expressible as the "energy of assembly"[24] -- *viz.*

$$-\sum_i^{all} q_i \int_{\vec{\mathbf{r}}_{\mathrm{ref}}}^{\vec{\mathbf{r}}_{\mathrm{i}}} \vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}})\; d\vec{\mathbf{r}} = -\sum_i^{all} q_i \int_{\vec{\mathbf{r}}_{\mathrm{ref}}}^{\vec{\mathbf{r}}=0} \vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}})\; d\vec{\mathbf{r}} + \sum_i^{all} q_i \int_{\vec{\mathbf{r}}=0}^{\vec{\mathbf{r}}_{\mathrm{i}}} \vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}})\; d\vec{\mathbf{r}} \qquad \text{[IV-15a]}$$

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[24] This is the work required to assemble the atom in the external field and of course ignore all inter-particle electrostatic interactions

which becomes for a neutral *atom* with charge centered at $\vec{\mathbf{r}} = 0$

$$U_{\text{int}} = \sum_{i}^{all} q_i \int_{\vec{\mathbf{r}}=0}^{\vec{\mathbf{r}}_{\mathrm{i}}} \vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}}\right) \; d\vec{\mathbf{r}} \qquad \text{[IV-15b]}$$

Electric fields of interest vary only slightly over an *atom* so that we should be able to expand the external field in a rapidly converging series as follows

$$\vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}}\right) = \vec{\mathbf{E}}_{\mathrm{T}}\left(0\right) + \left(\vec{\mathbf{r}}\;\vec{\nabla}\right) \vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}}\right)\Big|_{\vec{\mathbf{r}}=0} + \frac{1}{2!}\left(\vec{\mathbf{r}}\;\vec{\nabla}\right)^{2} \vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}}\right)\Big|_{\vec{\mathbf{r}}=0} + \ldots \qquad \text{[IV-16]}$$

Substituting this expansion into Equation [IV-15b] and integrating we obtain

$$U_{\text{int}} = -e \sum_{i}^{electrons} \vec{\mathbf{r}}_i \left\{ \left[1 + \frac{1}{2!}\left(\vec{\mathbf{r}}_i\;\vec{\nabla}\right) + \frac{1}{3!}\left(\vec{\mathbf{r}}_i\;\vec{\nabla}\right)^{2} + \cdots \right] \vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}}\right)\Big|_{\vec{\mathbf{r}}=0} \right\} \qquad \text{[IV-17a]}$$

Formally, we may cast this expansion for the interaction energy in the form

$$U_{\text{int}} = -e \sum_{i}^{electrons} \vec{\mathbf{r}}_i \left\{ \frac{\exp\left(\vec{\mathbf{r}}_i\;\vec{\nabla}\right) - 1}{\vec{\mathbf{r}}_i\;\vec{\nabla}} \; \vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}}\right)\Big|_{\vec{\mathbf{r}}=0} \right\} \qquad \text{[IV-17b]}$$

In the continuum picture, the interaction energy should be expressible in the form

$$U_{\text{int}} = \iiint \vec{\mathbf{E}}_{\mathrm{T}}\left(\vec{\mathbf{r}}\right) \; \vec{\mathbf{P}}\left(\vec{\mathbf{r}}\right) \, \mathrm{dV} \qquad \text{[IV-18]}$$

where $\vec{\mathbf{P}}\left(\vec{\mathbf{r}}\right)$ is the polarization density. It may be shown quite easily that Equations [IV-18] and [IV-17] are equivalent if the polarization density is expressed in the following

multipole expansion

$$\vec{\mathbf{P}}(\vec{\mathbf{r}}) = -e \sum_{i}^{electrons} \left\{ \vec{\mathbf{r}} - 1/2!\, \vec{\mathbf{r}}\left(\vec{\mathbf{r}} \quad \vec{\ }\right) + 1/3!\, \vec{\mathbf{r}}\left(\vec{\mathbf{r}} \quad \vec{\ }\right)^2 + \cdots \right\} \delta\left(\vec{\mathbf{r}} - \vec{\mathbf{r}}_i\right) \quad [\text{ IV-19 }]$$

With this background, we return to a consideration of the Hamiltonian in Equation [IV-14]. In what follows, we show that this Hamiltonian is transformed into a form which consistent with Equation [IV-17b] if we make an appropriate **gauge transformation** of the fields. In general, the gauge transformation

$$\begin{aligned} &\vec{\mathbf{A}}(\vec{\mathbf{r}}, t) \qquad && \vec{\mathbf{A}}(\vec{\mathbf{r}}, t) - \vec{\ }\chi(\vec{\mathbf{r}}, t) \\ &\phi(\vec{\mathbf{r}}, t) \qquad && \phi(\vec{\mathbf{r}}, t) + \frac{\partial}{\partial t}\chi(\vec{\mathbf{r}}, t) \end{aligned} \qquad [\text{ IV-20 }]$$

where $\chi(\vec{\mathbf{r}}, t)$ is an arbitrary scalar gauge function, leaves the electric and magnetic fields unchanged. Motivated by the discussion above, we choose the gauge function

$$\chi(\vec{\mathbf{r}}, t) = (1/e) \iiint \vec{\mathbf{A}}(\vec{\mathbf{r}}') \quad \vec{\mathbf{P}}(\vec{\mathbf{r}}')\ \mathrm{dV}' \qquad [\text{ IV-21 }]$$

where $\vec{\mathbf{P}}(\vec{\mathbf{r}})$ is the polarization density in the form shown in Equation [IV-19] and $\vec{\mathbf{A}}(\vec{\mathbf{r}})$ is the vector potential appearing in Equation [IV-14]. The impact of the transformation on the scalar potential term in Equation [IV-14] is easy to evaluate -- *viz*.

$$\phi(\vec{\mathbf{r}}_i, t) \qquad \phi(\vec{\mathbf{r}}_i, t) - \vec{\mathbf{r}}_i \left\{ \left[\frac{\exp\left(\vec{\mathbf{r}}_i \quad \vec{\ }\right) - 1}{\vec{\mathbf{r}}_i \quad \vec{\ }} \right] \vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}})\Big|_{\vec{\mathbf{r}}=0} \right\} \qquad [\text{ IV-22 }]$$

Dealing with the transformation

$$\vec{\mathbf{A}}(\vec{\mathbf{r}}_i, t) \qquad \vec{\mathbf{A}}(\vec{\mathbf{r}}_i, t) - (1/e)\iiint \vec{\ }_i\, \vec{\mathbf{A}}(\vec{\mathbf{r}})\ \vec{\mathbf{P}}(\vec{\mathbf{r}})\, \mathrm{dV} \qquad [\text{ IV-23 }]$$

is straightforward, but extremely tedious (and not very useful)! Substituting for $\vec{\mathbf{P}}(\vec{\mathbf{r}})$ from Equation [IV-19] and doing a lot of integrating by parts we could demonstrate that

$$\vec{\mathbf{A}}(\vec{\mathbf{r}}_i, t) \qquad -\mu_0\, \vec{\mathbf{r}}_i \times \left\{ \left[\frac{\exp(\vec{\mathbf{r}}_i\ \vec{\ })\,(\vec{\mathbf{r}}_i\ \vec{\ } - 1) + 1}{(\vec{\mathbf{r}}_i\ \vec{\ })^2} \right] \vec{\mathbf{H}}(\vec{\mathbf{r}})\Big|_{\vec{\mathbf{r}}=0} \right\} \qquad [\text{ IV-24 }]$$

Making use of these results, the complete Hamiltonian of a atom plus radiation field may be written

$$\begin{aligned} \mathcal{H} = {} & \frac{1}{2m}\sum_i \left\{ \vec{\mathcal{P}}_i - e\mu_0\, \vec{\mathbf{r}}_i \times \left\{ \left[\frac{\exp(\vec{\mathbf{r}}_i\ \vec{\ })\,(\vec{\mathbf{r}}_i\ \vec{\ } - 1) + 1}{(\vec{\mathbf{r}}_i\ \vec{\ })^2} \right] \vec{\mathbf{H}}(\vec{\mathbf{r}})\Big|_{\vec{\mathbf{r}}=0} \right\} \right\}^2 \\ & - \frac{1}{2}\, e \sum_i \phi(\vec{\mathbf{r}}_i) + \frac{1}{2}\, Ze\, \phi(0) + \frac{1}{2}\iiint \left[\varepsilon_0\, \vec{\mathbf{E}}_{\mathrm{T}}^{\,2} + \mu_0\, \vec{\mathbf{H}}^{\,2} \right] \mathrm{d}V \qquad [\text{ IV-25 }] \\ & + e \sum_i \vec{\mathbf{r}}_i \left\{ \left[\frac{\exp(\vec{\mathbf{r}}_i\ \vec{\ }) - 1}{\vec{\mathbf{r}}_i\ \vec{\ }} \right] \vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}})\Big|_{\vec{\mathbf{r}}=0} \right\} \end{aligned}$$

In practical terms, the ***useful*** complete Hamiltonian is written

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_A + \mathcal{H}_R + \mathcal{H}_I \\ &= \mathcal{H}_A + \mathcal{H}_R + \left(\mathcal{H}_{ED} + \mathcal{H}_{EQ} + \mathcal{H}_{MD} + \mathcal{H}_{NL} \right) \qquad [\text{ IV-26a }] \end{aligned}$$

where

$$\mathcal{H}_A = \sum_i \vec{\mathcal{P}}_i^{\,2}/2m - \frac{1}{2}\, e \sum_i \phi(\vec{\mathbf{r}}_i) + \frac{1}{2}\, Ze\, \phi(0) \qquad [\text{ IV-26b }]$$

$$\mathcal{H}_R = \frac{1}{2}\iiint\left[\varepsilon_0\,\vec{\mathbf{E}}_{\mathrm{T}}^{\,2} + \mu_0\,\vec{\mathbf{H}}^{\,2}\right]\mathrm{d}V \qquad \text{[IV-26c]}$$

$$\mathcal{H}_{ED} = e\sum_i \vec{\mathbf{r}}_i\ \ \vec{\mathbf{E}}_{\mathrm{T}}(0) = e\,\vec{\mathcal{D}}\ \ \vec{\mathbf{E}}_{\mathrm{T}}(0) \qquad \text{[IV-26d]}$$

$$\mathcal{H}_{EQ} = \frac{1}{2}\,e\sum_i \vec{\mathbf{r}}_i\ \left\{\left[\vec{\mathbf{r}}_i\ \ \vec{}\ \right]\vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}})\Big|_{\vec{\mathbf{r}}=0}\right\} = -\ \vec{}\ \ \overleftrightarrow{\mathcal{Q}}\ \ \vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}})\Big|_{\vec{\mathbf{r}}=0} \qquad \text{[IV-26e]}$$

$$\mathcal{H}_{MD} = \frac{e\,\mu_0}{2m}\sum_i\left[\vec{\mathbf{r}}_i\times\vec{\mathcal{P}}\right]\ \vec{\mathbf{H}}(0) = \frac{e\,\mu_0}{2m}\,\vec{\mathcal{M}}\ \ \vec{\mathbf{H}}(0) \qquad \text{[IV-26f]}$$

$$\mathcal{H}_{NL} = \frac{e^2\,\mu_0^{\ 2}}{8m}\sum_i\left[\vec{\mathbf{r}}_i\times\vec{\mathbf{H}}(0)\right]^2 \qquad \text{[IV-26g]}$$

If we take $\vec{\mathbf{r}}_i$ of the order of magnitude of the Bohr radius a_{B}, $\vec{\mathcal{M}}$ of order $\hbar$, and $\vec{}\ \vec{\mathbf{E}}_{\mathrm{T}}(\vec{\mathbf{r}})\Big|_{\vec{\mathbf{r}}=0}$ of order $|\vec{\mathbf{k}}|\,|\vec{\mathbf{E}}_{\mathrm{T}}(0)|$ then we may estimate the three linear interaction terms[25] -- *viz.*

$$\mathcal{H}_{ED} \approx \mathbf{E}_{\mathrm{T}}(0)\ e\ a_{\mathrm{B}} = \mathbf{E}_{\mathrm{T}}(0)\,\frac{4\pi\,\varepsilon_0\,\hbar^2}{m\,e}$$

$$\mathcal{H}_{EQ} \approx \mathbf{E}_{\mathrm{T}}(0)\,\frac{3\,e\,\hbar}{16\,m c}$$

$$\mathcal{H}_{MD} \approx \mathbf{H}(0)\,\frac{\mu_0\ e\,\hbar}{2\,m} = \mathbf{E}_{\mathrm{T}}(0)\,\frac{e\,\hbar}{2\,m c}$$

Further we note that $\dfrac{\mathcal{H}_{EQ}}{\mathcal{H}_{ED}}$ and $\dfrac{\mathcal{H}_{MD}}{\mathcal{H}_{ED}} \approx \dfrac{\mathrm{e}^2}{4\pi\,\varepsilon_0\,\hbar\,c} \approx \dfrac{1}{137}$!!

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[25] We assume here that $\hbar\,\omega = \hbar\,c\ k = e^2/2a_{\mathrm{B}} = \mathrm{R}_\infty/2$

V. PHOTON ABSORPTION AND EMISSION

"POOR MAN'S" SECOND QUANTIZATION OF MATERIAL SYSTEM:

In treating the complete quantum mechanical problem, it is useful to recast the material (atomic) Hamiltonian in terms of an appropriate set of ***creation*** and ***destruction*** operators. To that end we make the following definition

$$\mathcal{H}_A \left|x\right\rangle = \hbar\omega_x \left|x\right\rangle \qquad [\text{ V-1 }]$$

Using the ubiquitous identity operation $\sum_x |x\rangle\langle x| = 1$, we may write the material Hamiltonian in ***second quantized*** form -- *viz.*

$$\begin{aligned}\mathcal{H}_A &= \sum_x |x\rangle\langle x|\, \mathcal{H}_A \sum_y |y\rangle\langle y| = \sum_x \sum_y |x\rangle\, \hbar\omega_x \,\langle x|y\rangle\langle y| \\ &= \sum_x \hbar\omega_x \,|x\rangle\langle x|\end{aligned} \qquad [\text{ V-2 }]$$

In general, the operator $|x\rangle\langle y| \equiv b_x^\dagger\, b_y$ applied to any state $|z\rangle$ yields

$$|x\rangle\langle y|z\rangle = b_x^\dagger\, b_y\, |z\rangle = |x\rangle\, \delta_{yz} \qquad [\text{ V-3 }]$$

-- *i.e.* the operator changes a state **z** to a state **x** if the state is **y** otherwise it produces zero. In other words, the operator **destroys** the state **y** and **creates** a state **x**. The second quantization viewpoint is particularly useful in treating the interaction of a two-level material system with the radiation field. This case, is most conveniently formulate in two-vector notation with the use of ***Pauli spin matrices*** -- *viz.*

$$|a\rangle = \begin{bmatrix}1\\0\end{bmatrix} \quad \text{and} \quad |b\rangle = \begin{bmatrix}0\\1\end{bmatrix} \qquad [\text{ V-4a }]$$

$$\langle a| = \begin{bmatrix}1 & 0\end{bmatrix} \quad \text{and} \quad \langle b| = \begin{bmatrix}0 & 1\end{bmatrix} \qquad [\text{ V-4b }]$$

$$|a\rangle\langle b| = \begin{bmatrix}1\\0\end{bmatrix}\begin{bmatrix}0 & 1\end{bmatrix} = \begin{bmatrix}0 & 1\\0 & 0\end{bmatrix} = \sigma^{+}$$

[V-4c]

$$|b\rangle\langle a| = \begin{bmatrix}0\\1\end{bmatrix}\begin{bmatrix}1 & 0\end{bmatrix} = \begin{bmatrix}0 & 0\\1 & 0\end{bmatrix} = \sigma^{-}$$

$$|a\rangle\langle a| = \begin{bmatrix}1\\0\end{bmatrix}\begin{bmatrix}1 & 0\end{bmatrix} = \begin{bmatrix}1 & 0\\0 & 0\end{bmatrix}$$

[V-4d]

$$|b\rangle\langle b| = \begin{bmatrix}0\\1\end{bmatrix}\begin{bmatrix}0 & 1\end{bmatrix} = \begin{bmatrix}0 & 0\\0 & 1\end{bmatrix}$$

Consequently, the atomic Hamiltonian may be written

$$\mathcal{H}_A = \hbar\omega_a\begin{bmatrix}1 & 0\\0 & 0\end{bmatrix} + \hbar\omega_b\begin{bmatrix}0 & 0\\0 & 1\end{bmatrix}$$

[V-5a]

$$= \frac{1}{2}\hbar\left(\omega_a - \omega_b\right)\begin{bmatrix}1 & 0\\0 & -1\end{bmatrix} + \frac{1}{2}\hbar\left(\omega_a + \omega_b\right)\begin{bmatrix}1 & 0\\0 & 1\end{bmatrix}$$

and if we neglect the mean energy of the states

$$\mathcal{H}_A \quad \frac{1}{2}\hbar\omega_{ab}\begin{bmatrix}1 & 0\\0 & -1\end{bmatrix} = \frac{1}{2}\hbar\omega_{ab}\,\sigma^{z}$$

[V-5b]

and the electric dipole interaction Hamiltonian becomes

$$\begin{aligned}\mathcal{H}_{ED} &= e\,\vec{\mathcal{D}}\;\;\vec{\mathbf{E}}_{\mathrm{T}}(0)\\ &= \Big[\,|a\rangle\langle a|+|b\rangle\langle b|\,\Big]\Big[e\,\vec{\mathcal{D}}\Big]\Big[\,|a\rangle\langle a|+|b\rangle\langle b|\,\Big]\;\vec{\mathbf{E}}_{\mathrm{T}}(0)\\ &= \Big\{\,|a\rangle\langle a|\Big[e\,\vec{\mathcal{D}}\Big]|b\rangle\langle b|+|b\rangle\langle b|\Big[e\,\vec{\mathcal{D}}\Big]|a\rangle\langle a|\,\Big\}\;\vec{\mathbf{E}}_{\mathrm{T}}(0)\\ &= \Big\{e\langle a|\,\vec{\mathcal{D}}\,|b\rangle\,\sigma^{+}+e\langle b|\,\vec{\mathcal{D}}\,|a\rangle\,\sigma^{-}\Big\}\;\vec{\mathbf{E}}_{\mathrm{T}}(0)\\ &= -\Big\{\;\vec{\ }\;\sigma^{+}+\;\vec{\ }\;\;\sigma^{-}\Big\}\;\vec{\mathbf{E}}_{\mathrm{T}}(0)\end{aligned} \qquad [\text{ V-6 }]$$

From Equation [II-24a] in this lecture set we can write

$$\vec{\mathbf{E}}_{\mathrm{T}}(0)\quad \sum_{\{l\}}\sum_{\sigma=1}^{2}\hat{\mathbf{e}}_{\{l\}\,\sigma}\,\mathcal{E}_{\{l\}}\Big\{\,i\,a_{\{l\}\,\sigma}(t)\,\exp\Big[i\,\vec{\mathbf{k}}_{\{l\}}\;\;\vec{\mathbf{r}}_{A}\Big]-i\,a^{\dagger}_{\{l\}\,\sigma}(t)\,\exp\Big[-i\,\vec{\mathbf{k}}_{\{l\}}\;\;\vec{\mathbf{r}}_{A}\Big]\Big\} \qquad [\text{ V-7 }]$$

where $\mathcal{E}_{\{\}} = \sqrt{\dfrac{\hbar\omega_{\{l\}}}{2\varepsilon_0\,V}}$ is the so called the ***electric field per photon*** and $\vec{\mathbf{r}}_A$ is the location of the center of the atom under consideration. Thus Equation [V-6] may be written quite generally for a two level atom as

$$\begin{aligned}\mathcal{H}_{ED} &= \Big\{e\langle a|\,\vec{\mathcal{D}}\,|b\rangle\,\sigma^{+}+e\langle b|\,\vec{\mathcal{D}}\,|a\rangle\,\sigma^{-}\Big\}\\ &\qquad\times\sum_{\{l\}\,]}\sum_{s=1}^{2}\hat{\mathbf{e}}_{\{l\}\,s}\,\mathcal{E}_{\{\}}\Big\{\,i\,a_{\{l\}\,s}(t)\,\exp\Big[i\,\vec{\mathbf{k}}_{\{\}}\;\;\vec{\mathbf{r}}_{A}\Big]-i\,a^{\dagger}_{\{l\}\,s}(t)\,\exp\Big[-i\,\vec{\mathbf{k}}_{\{l\}}\;\;\vec{\mathbf{r}}_{A}\Big]\Big\} \qquad [\text{ V-8 }]\\ &= \hbar\sum_{\{l\}}\sum_{s=1}^{2}\Big\{g_{\{l\}\,s}\,\sigma^{+}+g_{\{l\}\,s}\,\sigma^{-}\Big\}\Big\{\,i\,a_{\{l\}\,s}(t)\,\exp\Big[i\,\vec{\mathbf{k}}_{\{l\}}\;\;\vec{\mathbf{r}}_{A}\Big]+adj.\Big\}\end{aligned}$$

where the coupling constant is given by

$$g_{\{\ell\}\,s} = \hbar^{-1}\, e\, \mathcal{E}_{\{l\}}\ \vec{\ }_{A}\ \ \hat{\mathbf{e}}_{\{\ell\}\,s} = e\sqrt{\frac{\omega_{\{\ell\}}}{2\,\hbar\,\varepsilon_0}}\ \langle a|\,\vec{\mathcal{D}}\,|b\rangle\ \ \hat{\mathbf{e}}_{\{\ell\}\,s} \qquad [\text{ V-9 }]$$

Interaction of a Two-level Atom and a Single Mode Field -- Rabi Flopping:

Let us first consider the interaction of a two-level system with a single photon state to make contact with the discussion in Section III of the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*. Equation [V-8] then reduces to

$$\mathcal{H}_{ED} = i\,\hbar\left\{g_{\vec{\mathbf{k}}}\,\sigma^{+} + g_{\vec{\mathbf{k}}}\,\sigma^{-}\right\}\left\{\,a_{\vec{\mathbf{k}}}(t)\ \exp\!\left[i\,\vec{\mathbf{k}}\ \ \vec{\mathbf{r}}_A\right] - a^{\dagger}_{\vec{\mathbf{k}}}(t)\ \exp\!\left[-i\,\vec{\mathbf{k}}\ \ \vec{\mathbf{r}}_A\right]\right\} \qquad [\text{ V-10a }]$$

where $g_{\vec{\mathbf{k}}} = \hbar^{-1}\, e\, \mathcal{E}_{\vec{\mathbf{k}}}\ \vec{\ }_{A}\ \ \hat{\mathbf{e}}_{\vec{\mathbf{k}}} = e\sqrt{\frac{\omega_{\vec{\mathbf{k}}}}{2\,\hbar\,\varepsilon_0}}\ \langle a|\,\vec{\mathcal{D}}\,|b\rangle\ \ \hat{\mathbf{e}}_{\vec{\mathbf{k}}}$. If we neglect any inter-atomic interference effects by taking $\vec{\mathbf{r}}_A = 0$, we may simplify Equation [V-10a] to

$$\begin{aligned}\mathcal{H}_{ED} &= \hbar\left\{\ g_{\vec{\mathbf{k}}}\,\sigma^{+} + g_{\vec{\mathbf{k}}}\,\sigma^{-}\ \right\}\left\{\ i\,a_{\vec{\mathbf{k}}}(t) - i\,a^{\dagger}_{\vec{\mathbf{k}}}(t)\ \right\}\\ &= \hbar\left\{\ g_{\vec{\mathbf{k}}}\,\sigma^{+} + g_{\vec{\mathbf{k}}}\,\sigma^{-}\ \right\}\left\{\ i\,a_{\vec{\mathbf{k}}}(t) + adj.\right\}\end{aligned} \qquad [\text{ V-10b }]$$

Thus, we may then write the complete effective Hamiltonian of the composite system as

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I = \frac{1}{2}\hbar\omega_{ab}\ \sigma^{z} + \hbar\omega_{\vec{\mathbf{k}}}\ a^{\dagger}a + \hbar\left\{\ g_{\vec{\mathbf{k}}}\,\sigma^{+} + g_{\vec{\mathbf{k}}}\,\sigma^{-}\ \right\}\left\{\ i\,a_{\vec{\mathbf{k}}}(t) - i\,a^{\dagger}_{\vec{\mathbf{k}}}(t)\right\} \qquad [\text{ V-11a }]$$

In the **rotating wave approximation** this reduces to

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I \approx \frac{1}{2}\hbar\omega_{ab}\ \sigma^{z} + \hbar\omega_{\vec{\mathbf{k}}}\ a^{\dagger}a + \hbar\left\{i\ g_{\vec{\mathbf{k}}}\ a\ \sigma^{+} - i\ g_{\vec{\mathbf{k}}}\ a^{\dagger}\ \sigma^{-}\right\} \qquad [\text{ V-11b }]$$

In the spirit of the discussion in the Section VII, *Semiconductor Photonics* in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*, this effective Hamiltonian may be adapted to provide a fully quantum mechanical treatment of optical interactions in semiconductors.[26]

"DRESSED" ATOMIC STATES:

We know that the unperturbed Hamiltonian satisfies the following eigenvalue equations

$$\begin{aligned}\mathcal{H}_0\,|a\;n\rangle &= \hbar\left[\frac{1}{2}\omega_{ab} + n\,\omega_{\vec{\mathbf{k}}}\right]|a\;n\rangle \\ \mathcal{H}_0\,|b\;n\rangle &= \hbar\left[-\frac{1}{2}\omega_{ab} + n\,\omega_{\vec{\mathbf{k}}}\right]|b\;n\rangle\end{aligned} \qquad [\text{ V-12 }]$$

-- where $|a\;n\rangle \equiv |a\rangle|n_{\vec{\mathbf{k}}}\rangle$ and $|b\;n\rangle \equiv |b\rangle|n_{\vec{\mathbf{k}}}\rangle$ -- and the electric dipole perturbation couples the states $|a\;n\rangle$ and $|b\;(n+1)\rangle$. It is useful to resolve the complete

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[26] In *Semiconductor Photonics* we noted that Chow, Koch and Sargent in their *Semiconductor-Laser Physics* (Springer-Verlag - 1994) treat semiconductor problems in terms of the following semiclassical Hamiltonian for an inhomogeneous two-level system:

$$\mathcal{H}_{\text{eff}} = \sum_{\vec{\mathbf{k}}}\left\{\left[\mathcal{E}_g + \frac{\hbar^2\,k^2}{2m_{ec}}\right]a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}} + \left[\frac{\hbar^2\,k^2}{2m_{hv}}\right]b^{\dagger}_{-\vec{\mathbf{k}}}\,b_{-\vec{\mathbf{k}}} - \left[\mu_{\vec{\mathbf{k}}}\,a^{\dagger}_{\vec{\mathbf{k}}}\,b^{\dagger}_{-\vec{\mathbf{k}}} + \mu_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\,b_{-\vec{\mathbf{k}}}\right]\mathrm{E}(z,t)\right\}$$

where $\{a^{\dagger}_{\vec{\mathbf{k}}}, a_{\vec{\mathbf{k}}}\}$ and $\{b^{\dagger}_{\vec{\mathbf{k}}}, b_{\vec{\mathbf{k}}}\}$ are, respectively, electron and hole $\{$**creation, destruction**$\}$ operators. $\mu_{\vec{\mathbf{k}}}$ is the dipole matrix element between vertical states in the valence and conduction bands. In the fully quantal treatment the effective Hamiltonian becomes

$$\mathcal{H}_{\text{eff}} = \sum_{\vec{\mathbf{k}}}\left\{\left[\mathcal{E}_g + \frac{\hbar^2\,k^2}{2m_{ec}}\right]a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}} + \left[\frac{\hbar^2\,k^2}{2m_{hv}}\right]b^{\dagger}_{-\vec{\mathbf{k}}}\,b_{-\vec{\mathbf{k}}} - \hbar\left[g_{\vec{\mathbf{k}}}\,a^{\dagger}_{\vec{\mathbf{k}}}\,b^{\dagger}_{-\vec{\mathbf{k}}}\,a + g_{\vec{\mathbf{k}}}\,a^{\dagger}\,a_{\vec{\mathbf{k}}}\,b_{-\vec{\mathbf{k}}}\right]\right\}$$

where $g_{\vec{\mathbf{k}}} = \sqrt{2}\,\mu_{\vec{\mathbf{k}}}\,\mathcal{E}_{\omega_r}\,\sin(k_r\,z_0)/\hbar$

Hamiltonian into a sum of component Hamiltonians $\mathcal{H} = \sum_n \mathcal{H}_n$ where the component $\mathcal{H}_n$'s act only within the $\{|a\ n\rangle,\ |b\ (n+1)\rangle\}$ coupled manifold of states and can be written[27]

$$\mathcal{H}_n \quad \hbar\left(n+\tfrac{1}{2}\right)\omega_{\bar{\mathbf{k}}}\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}+\frac{\hbar}{2}\begin{bmatrix}\delta\omega & 2\,i\,g_{\bar{\mathbf{k}}}\,a\\ -2\,i g_{\bar{\mathbf{k}}}\,a^{\dagger} & -\delta\omega\end{bmatrix}$$

$$\hbar\left(n+\tfrac{1}{2}\right)\omega_{\bar{\mathbf{k}}}\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}+\frac{\hbar}{2}\begin{bmatrix}\delta\omega & 2\,i\,g_{\bar{\mathbf{k}}}\sqrt{n+1}\\ -2\,i g_{\bar{\mathbf{k}}}\sqrt{n+1} & -\delta\omega\end{bmatrix} \qquad [\text{ V-13 }]$$

where $\delta\omega = \omega_{ab} - \omega_{\bar{\mathbf{k}}}$. The second term in this equation is of the same form as the coupling matrix in Equation [III-8c] of the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory* where the semiclassical Rabi frequency $\Omega_{\mathbf{o}}^{\mathbf{R}} = \quad E_0/\hbar$ is replaced by its quantum equivalent -- *viz.* $2\,i\,g_{\bar{\mathbf{k}}}\sqrt{n+1}$. Diagonalizing this matrix we find the eigenvalues of the so call ***dressed atomic states*** -- *viz.*[28]

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[27] In particular, using the **identity operator** -- *i.e.*,. $I = \sum_\eta |\eta\rangle\langle\eta|$

$$\mathcal{H}_n \quad \left\{|an\rangle\langle an| + |b(n+1)\rangle\langle b(n+1)|\right\}\ \mathcal{H}\ \left\{|an\rangle\langle an| + |b(n+1)\rangle\langle b(n+1)|\right\}$$
$$= |an\rangle\langle an|\ \langle an|\ \mathcal{H}\ |an\rangle + |an\rangle\langle b(n+1)|\ \langle an|\ \mathcal{H}\ |b(n+1)\rangle$$
$$+\,|b(n+1)\rangle\langle an|\ \langle b(n+1)|\ \mathcal{H}\ |an\rangle + |b(n+1)\rangle\langle b(n+1)|\ \langle b(n+1)|\ \mathcal{H}\ |b(n+1)\rangle$$

[28] It should be noted that the rotation matrix

$$= \begin{bmatrix}\cos\theta_n & -\sin\theta_n\\ \sin\theta_n & \cos\theta_n\end{bmatrix}$$

diagonalizes the Hamiltonian in Equation [V-13] through the transformation $\quad \mathcal{H}_n \quad ^{-1}$. Further, relates the ***dressed*** and ***bare*** probability amplitudes as

$$\begin{aligned} E_{1n} &= \hbar\left(n+\tfrac{1}{2}\right)\omega_{\vec{\mathbf{k}}} + \tfrac{1}{2}\,\hbar\Omega_n^{\mathbf{R}} \\ E_{2n} &= \hbar\left(n+\tfrac{1}{2}\right)\omega_{\vec{\mathbf{k}}} - \tfrac{1}{2}\,\hbar\Omega_n^{\mathbf{R}} \end{aligned} \qquad [\text{ V-14 }]$$

(see energy level diagram below) where $\Omega_n^{\mathbf{R}} \equiv \sqrt{\delta\omega^2 + 4\left|g_{\vec{\mathbf{k}}}\right|^2(n+1)}$ is the **quantized field generalization** of the **Rabi flopping frequency.**

The dressed eigenstates are given by

$$\begin{aligned} |1n\rangle &= \sin\theta_n\,|a\;n\rangle + \cos\theta_n\,|b\;(n+1)\rangle \\ |2n\rangle &= \cos\theta_n\,|a\;n\rangle - \sin\theta_n\,|b\;(n+1)\rangle \end{aligned} \qquad [\text{ V-15a }]$$

where

$$\cos\theta_n = \frac{\Omega_n^{\mathbf{R}} - \delta\omega}{\sqrt{\left(\Omega_n^{\mathbf{R}} - \delta\omega\right)^2 + 4\left|g_{\vec{\mathbf{k}}}\right|^2(n+1)}} \qquad [\text{ V-15b }]$$

and

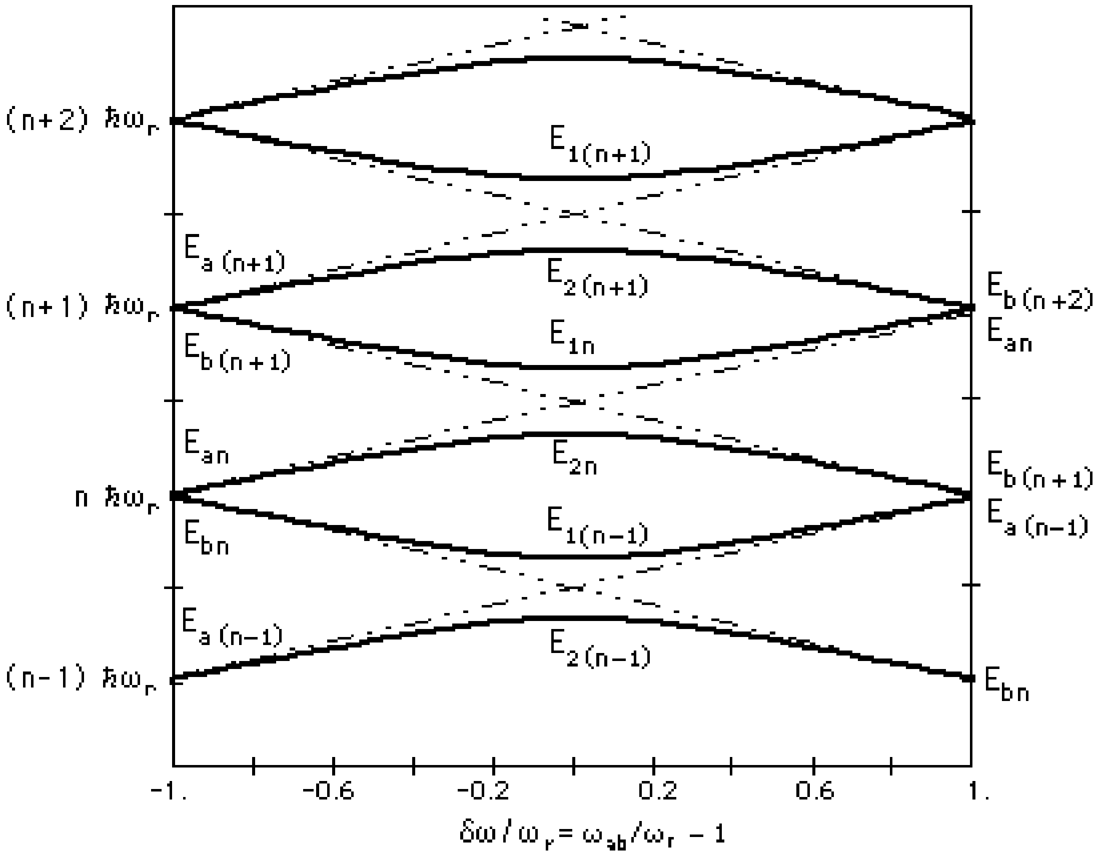
$$\sin\theta_n = \frac{2g_r\sqrt{n+1}}{\sqrt{\left(\Omega_n^{\mathbf{R}} - \delta\omega\right)^2 + 4\left|g_{\vec{\mathbf{k}}}\right|^2(n+1)}} \qquad [\text{ V-15c }]$$

$$\begin{bmatrix} C_{2n}(t) \\ C_{1n}(t) \end{bmatrix} = \begin{bmatrix} C_{an}(t) \\ C_{b(n+1)}(t) \end{bmatrix}$$

where

$$\begin{aligned} |\psi\rangle &= \sum_n \left[C_{an}(t)\,|a\;n\rangle + C_{b(n+1)}(t)\,|b\;(n+1)\rangle\right] \\ &= \sum_n \left[C_{1n}(t)\,|1n\rangle + C_{2n}(t)\,|2n\rangle\right] \end{aligned}$$

DRESSED **ENERGY LEVELS OF TWO-STATE ATOM**



The time evolution of a state is directly represented in terms of these dressed states -- *viz*.

$$
\begin{aligned}
\left|\psi(t)\right\rangle &= \exp\left(-i\,\mathcal{H}\;\, t/\hbar\right)\left|\psi(0)\right\rangle \\
&= \sum_{n=0}^{\infty}\sum_{s=1}^{2}\exp\left(-i E_{sn}\; t/\hbar\right)\left|s\,n\right\rangle\left\langle s\,n\,\middle|\,\psi(0)\right\rangle \qquad \text{[V-16a]} \\
&= \sum_{n=0}^{\infty}\exp\left(-i\left[n+\tfrac{1}{2}\right]\omega_{\bar{\mathbf{k}}}\; t\right)\left\{\;\exp\left(-\tfrac{1}{2}\, i\,\Omega_n^{\mathbf{R}}\;\, t\right)\left|1\;n\right\rangle C_{1n}(0)+\exp\left(\tfrac{1}{2}\; i\,\Omega_n^{\mathbf{R}}\;\, t\right)\left|2\;n\right\rangle C_{2n}(0)\;\right\}
\end{aligned}
$$

or more explicitly

$$
\begin{bmatrix} C_{2n}(t) \\ C_{1n}(t) \end{bmatrix} = \begin{bmatrix} \exp\left(\tfrac{1}{2}\; i\,\Omega_n^{\mathbf{R}}\;\, t\right) & 0 \\ 0 & \exp\left(-\tfrac{1}{2}\; i\,\Omega_n^{\mathbf{R}}\;\, t\right) \end{bmatrix} \begin{bmatrix} C_{2n}(0) \\ C_{1n}(0) \end{bmatrix} \qquad \text{[V-16b]}
$$

Therefore, the **flopping** of the undressed states is given by

$$
\begin{bmatrix} C_{an}(t) \\ C_{b(n+1)}(t) \end{bmatrix} = {}^{-1}\begin{bmatrix} \exp\left(\tfrac{1}{2}\; i\,\Omega_n^{\mathbf{R}}\;\, t\right) & 0 \\ 0 & \exp\left(-\tfrac{1}{2}\; i\,\Omega_n^{\mathbf{R}}\;\, t\right) \end{bmatrix} \begin{bmatrix} C_{an}(0) \\ C_{b(n+1)}(0) \end{bmatrix}
$$

$$
= \begin{bmatrix} \cos\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right)+i\,\sin\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right)\,\cos 2\theta_n & -i\,\sin\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right)\,\sin 2\theta_n \\ -i\,\sin\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right)\,\sin 2\theta_n & \cos\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right)-i\,\sin\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right)\,\cos 2\theta_n \end{bmatrix} \begin{bmatrix} C_{an}(0) \\ C_{b(n+1)}(0) \end{bmatrix} \qquad \text{[V-17]}
$$

$$
= \begin{bmatrix} \cos\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right)-i\left(\delta\omega/\Omega_n^{\mathbf{R}}\right)\,\sin\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right) & -i\left(2g_r\sqrt{n+1}/\Omega_n^{\mathbf{R}}\right)\,\sin\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right) \\ -i\left(2g_r\sqrt{n+1}/\Omega_n^{\mathbf{R}}\right)\,\sin\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right) & \cos\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right)+i\left(\delta\omega/\Omega_n^{\mathbf{R}}\right)\,\sin\left(\tfrac{1}{2}\,\Omega_n^{\mathbf{R}}\;\, t\right) \end{bmatrix} \begin{bmatrix} C_{an}(0) \\ C_{b(n+1)}(0) \end{bmatrix}
$$

Perhaps the most revealing application of the this result is for the case of a resonant coupled system -- *i.e.* $\delta\omega = 0$ -- which is prepared so that $C_{b(n+1)}(0) = 0$. In this instance, Equation [V-17] yields

$$\begin{aligned} \left|C_{an}(t)\right|^2 &= \cos^2\left[g_{\mathbf{k}}\sqrt{n+1}\;t\right]\left|C_{an}(0)\right|^2 \\ \left|C_{b(n+1)}(t)\right|^2 &= \sin^2\left[g_{\mathbf{k}}\sqrt{n+1}\;t\right]\left|C_{an}(0)\right|^2 \end{aligned} \qquad [\text{ V-18 }]$$

which clearly exhibits the simplest manifestation of ***spontaneous emission*** -- *i.e.* Rabi flopping in the absence of an applied field!!!

Interaction of a Two-level Atom and a Multi Mode Field -- Spontaneous Emission:

To broaden (make more realistic) our treatment of spontaneous emission we return to Equation [V-8] to include the interaction with many modes with a two-level atom -- *viz.*

$$\mathcal{H}_{ED} = \hbar \sum_{\{l\}} \sum_{s=1}^{2} \left\{ g_{\{l\}s}\,\sigma^{+} + g_{\{l\}s}\,\sigma^{-} \right\} \left\{ i\, a_{\{l\}s}(t) \exp\left[i\,\vec{\mathbf{k}}_{\{l\}}\;\;\vec{\mathbf{r}}_A\right] + adj. \right\} \qquad [\text{ V-8' }]$$

This equation may be easily generalized to encompass multi-level material systems.[29]

If we are dealing with situation in which the locations of the atoms are **uncorrelated** we may, for simplicity, dispense with the $\exp\left[\pm i\,\vec{\mathbf{k}}_{\{\ell\}}\;\;\vec{\mathbf{r}}_0\right]$ factors -- *i.e.* we will neglect, for the present, any possible ***interference effects*** -- and write

---

[29] For a multilevel atomic system the effective interaction Hamiltonian in second quantized form becomes

$$\mathcal{H}_{ED} = e \sum_{\alpha,\beta} \sum_{\{l\}} \sum_{s=1}^{2} \sqrt{\frac{\hbar\omega_{\{l\}}}{2\varepsilon_0}} \left\{ \hat{\mathbf{e}}_{\{l\}s}\;\left\langle \alpha \right| \mathcal{D} \left| \beta \right\rangle \right\} \left\{ i\, a_{\{l\}s}(t) \exp\left[i\,\vec{\mathbf{k}}_{\{l\}}\;\;\vec{\mathbf{r}}_0\right] + adj. \right\} \left|\alpha\right\rangle\left\langle\beta\right|$$

$$\mathcal{H}_{ED} = \hbar \sum_{\{\ell\}} \sum_{s=1}^{2} \left[g_{\{\ell\} s}\, \sigma^{+} + g_{\{\ell\} s}\, \sigma^{-} \right] \left[i\, a_{\{\ell\} s}(t) - i\, a^{\dagger}_{\{\ell\} s}(t) \right] \qquad [\text{ V-19 }]$$

Transforming to the Schrödinger picture and taking the unperturbed ground state $|g\rangle = |b\rangle|0\rangle|0\rangle\cdots|0\rangle$ as the zero energy reference point we may write the complete effective Hamiltonian as

$$\begin{aligned} \mathcal{H} &= \hbar\omega_{ab}\, \sigma^{+}\, \sigma^{-} \\ &+ \sum_{\{\ell\}} \sum_{s=1}^{2} \left\{ \hbar\omega_{\{\ell\}}\, a^{\dagger}_{\{\ell\} s}\, a_{\{\ell\} s} + \hbar \left[g_{\{\ell\} s}\, \sigma^{+} + g_{\{\ell\} s}\, \sigma^{-} \right] \left[i\, a_{\{\ell\} s} - i\, a^{\dagger}_{\{\ell\} s} \right] \right\} . \end{aligned} \qquad [\text{ V-20a }]$$

If we include only **energy-conserving** terms -- *i.e.* in the rotating wave approximation --

$$\begin{aligned} \mathcal{H} &= \hbar\omega_{ab}\, \sigma^{+}\, \sigma^{-} \\ &+ \sum_{\{\ell\}} \sum_{s=1}^{2} \left\{ \hbar\omega_{\{\ell\}}\, a^{\dagger}_{\{\ell\} s}\, a_{\{\ell\} s} + \hbar \left[i g_{\{\ell\} s}\, \sigma^{+}\, a_{\{\ell\} s} - i g_{\{\ell\} s}\, \sigma^{-}\, a^{\dagger}_{\{\ell\} s} \right] \right\} . \end{aligned} \qquad [\text{ V-20b }]$$

It is important to note that, in general, the interaction terms in this Hamiltonian include contributions from the coupling of the material system (atom) to any externally excited mode(s) (the incident electromagnetic field) and to all available electromagnetic cavity modes. For the present, we ignore the coupling to externally excited modes: we treat the external interaction later as a perturbation. Our goal at this point is to diagonalize the Hamiltonian of the complete unperturbed system which may be written

$$\hbar^{-1}\mathcal{H} = \begin{bmatrix}\sigma^{+} & a_1^{\dagger} & a_2^{\dagger} & \cdots & a_N^{\dagger}\end{bmatrix}\begin{bmatrix}\omega_0 & ig_1 & ig_2 & \cdots & i\,g_N \\ -ig_1 & \omega_1 & 0 & \cdots & 0 \\ -ig_2 & 0 & \omega_2 & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ -i\,g_N & 0 & 0 & \cdots & \omega_N\end{bmatrix}\begin{bmatrix}\sigma^{-} \\ a_1 \\ a_2 \\ \vdots \\ a_N\end{bmatrix} . \qquad [\text{ V-21 }]$$

$$= \vec{\mathbf{a}}^{\dagger}\left(\overleftrightarrow{\omega}+\overleftrightarrow{\mathbf{g}}\right)\vec{\mathbf{a}}$$

We are looking for the eigenstates and eigenvalues of this Hamiltonian which we write as

$$\mathcal{H}\left|s\right\rangle = \hbar\lambda_s\left|s\right\rangle . \qquad [\text{ V-22 }]$$

Since the square matrix is Hermitian, it is possible, **in principle**, to diagonalize it via a unitary transformation of the form

$$\overleftrightarrow{\mathbf{U}}\left(\overleftrightarrow{\omega}+\overleftrightarrow{\mathbf{g}}\right)\overleftrightarrow{\mathbf{U}}^{-1} = \overleftrightarrow{\lambda} \qquad [\text{ V-23 }]$$

where $\overleftrightarrow{\lambda}$ is a diagonal matrix whose elements are the $N+1$ eigenvalues of the ***dressed*** states of the coupled system. If we define the row vector $\vec{\mu}^{\dagger} \equiv \vec{\mathbf{a}}^{\dagger}\,\overleftrightarrow{\mathbf{U}}^{-1}$ and the column vector $\vec{\mu} \equiv \overleftrightarrow{\mathbf{U}}\,\vec{\mathbf{a}}$ then Equation [V-22] becomes

$$\hbar^{-1}\mathcal{H} = \vec{\mathbf{a}}^{\dagger}\left(\overleftrightarrow{\omega}+\overleftrightarrow{\mathbf{g}}\right)\vec{\mathbf{a}} = \vec{\mu}^{\dagger}\,\overleftrightarrow{\lambda}\,\vec{\mu} = \sum_{t=1}^{N+1}\lambda_t\,\mu_t^{\dagger}\mu_t \qquad [\text{ V-24 }]$$

For consistency, $\left\langle r\right|\mu_t^{\dagger}\mu_t\left|s\right\rangle = \delta_{rs}\,\delta_{ts}$ and, hence, $\vec{\mu}^{\dagger}$ and $\vec{\mu}$ are, respectively, paired creation and destruction operators in the sense of the operators defined in Equation [V-3] above -- *i.e.* $\left|s\right\rangle = \mu_s^{\dagger}\left|\mathcal{g}\right\rangle$ and $\left|\mathcal{g}\right\rangle = \mu_s\left|s\right\rangle$. From Equation [V-23] we may write

$$\overleftrightarrow{\mathbf{U}}\ \left(\overleftrightarrow{\omega}+\overleftrightarrow{\mathbf{g}}\right)=\overleftrightarrow{\lambda}\ \ \overleftrightarrow{\mathbf{U}} \qquad [\text{ V-25a }]$$

which can be written explicitly as

$$\begin{bmatrix} \mathrm{U}_{11} & \mathrm{U}_{12} & \mathrm{U}_{13} & \cdots & \mathrm{U}_{1(N+1)} \\ \mathrm{U}_{21} & \mathrm{U}_{22} & \mathrm{U}_{23} & \cdots & \mathrm{U}_{2(N+1)} \\ \mathrm{U}_{31} & \mathrm{U}_{32} & \mathrm{U}_{33} & \cdots & \mathrm{U}_{3(N+1)} \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ \mathrm{U}_{(N+1)1} & \mathrm{U}_{(N+1)2} & \mathrm{U}_{(N+1)3} & \cdots & \mathrm{U}_{(N+1)(N+1)} \end{bmatrix} \begin{bmatrix} \omega_0 & ig_1 & i\,g_2 & \cdots & i\,g_N \\ -ig_1 & \omega_1 & 0 & \cdots & \\ -ig_2 & 0 & \omega_2 & \cdots & \\ \vdots & \vdots & \vdots & \vdots & \\ -ig_N & 0 & 0 & \cdots & \omega_N \end{bmatrix}$$

$$= \begin{bmatrix} \lambda_1\ \mathrm{U}_{11} & \lambda_1\ \mathrm{U}_{12} & \lambda_1\ \mathrm{U}_{13} & \cdots & \lambda_1\ \mathrm{U}_{1(N+1)} \\ \lambda_2\ \mathrm{U}_{21} & \lambda_2\ \mathrm{U}_{22} & \lambda_2\ \mathrm{U}_{23} & \cdots & \lambda_2\ \mathrm{U}_{2(N+1)} \\ \lambda_3\ \mathrm{U}_{31} & \lambda_3\mathrm{U}_{32} & \lambda_3\mathrm{U}_{33} & \cdots & \lambda_3\mathrm{U}_{3(N+1)} \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ \lambda_{(N+1)}\mathrm{U}_{(N+1)1} & \lambda_{(N+1)}\mathrm{U}_{(N+1)2} & \lambda_{(N+1)}\mathrm{U}_{(N+1)3} & \cdots & \lambda_{(N+1)}\mathrm{U}_{(N+1)(N+1)} \end{bmatrix} \qquad [\text{ V-25b }]$$

Expanding out the matrix product on the left we see that

$$\mathrm{U}_{s1}\,\omega_0 - i\sum_{\mathrm{t}}\mathrm{U}_{s(t+1)}\ g_t = \lambda_s\ \mathrm{U}_{s1} \qquad [\text{ V-26a }]$$

for elements **in the first column** and

$$i\,\mathrm{U}_{s1}\,g_{t-1} + \mathrm{U}_{st}\ \omega_{t-1} = \lambda_s \mathrm{U}_{st} \qquad s \neq t \qquad [\text{ V-26b }]$$

for elements **not in the first column**. Hence

$$\mathrm{U}_{st} = i\ \mathrm{U}_{s1}\ g_{t-1}\big/\left(\lambda_s - \omega_{t-1}\right) \qquad s \neq t \qquad [\text{ V-26c }]$$

and all elements of the unitary matrix can be expressed in terms of the first-column elements as

$$\overleftrightarrow{\mathbf{U}} = \begin{bmatrix} U_{11} & \dfrac{i\,U_{11}\,g_1}{\lambda_1-\omega_1} & \dfrac{i\,U_{11}\,g_2}{\lambda_1-\omega_2} & \cdots & \dfrac{i\,U_{11}\,g_N}{\lambda_1-\omega_N} \\ U_{21} & \dfrac{i\,U_{21}\,g_1}{\lambda_2-\omega_1} & \dfrac{i\,U_{21}\,g_2}{\lambda_2-\omega_2} & \cdots & \dfrac{i\,U_{21}\,g_N}{\lambda_2-\omega_N} \\ U_{31} & \dfrac{i\,U_{31}\,g_1}{\lambda_3-\omega_1} & \dfrac{i\,U_{31}\,g_2}{\lambda_3-\omega_2} & \cdots & \dfrac{i\,U_{31}\,g_N}{\lambda_3-\omega_N} \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ U_{(N+1)1} & \dfrac{i\,U_{(N+1)1}\,g_1}{\lambda_{(N+1)}-\omega_1} & \dfrac{i\,U_{(N+1)1}\,g_2}{\lambda_{(N+1)}-\omega_2} & \cdots & \dfrac{i\,U_{(N+1)1}\,g_N}{\lambda_{(N+1)}-\omega_N} \end{bmatrix} \qquad [\text{ V-27 }]$$

Quite generally the columns of any unitary matrix satisfy an orthonormal condition -- *viz*.

$$\sum_t U_{tr}\;U_{ts} = \delta_{rs} \qquad [\text{ V-28a }]$$

so the normalization of the first column gives

$$\sum_t \left| U_{ts} \right|^2 = 1 \qquad [\text{ V-28b }]$$

and the orthogonality of the first column with any other column yields

$$\sum_t \frac{\left|U_{t1}\right|^2}{\lambda_t-\omega_q} = 0 \qquad [\text{ V-29c }]$$

for each one of the cavity frequencies ω_q.

Further we may multiply Equation [V-28c] by a product of factors

$$\sum_t \frac{\left|U_{t1}\right|^2}{\omega_q - \lambda_t} \prod_s \left(\omega_q - \lambda_s\right) = 0 \qquad \text{[V-28d]}$$

It is obvious from this expression the ω_q.'s are roots of the left side of the equation if the ω_q.'s are replaced by ω . Thus

$$\sum_t \frac{\left|U_{t1}\right|^2}{\omega - \lambda_t} \prod_s \left(\omega - \lambda_t\right) = \prod_q \left(\omega - \omega_q\right) \qquad \text{[V-28e]}$$

Finally, we see that

$$\sum_t \frac{\left|U_{t1}\right|^2}{\lambda_t - \omega} = \frac{\prod_q \left(\omega - \omega_q\right)}{\prod_s \left(\omega - \lambda_s\right)} \qquad \text{[V-29]}$$

We can make use of this expression to obtain the time varying polarization induced by an externally excited field. The Hamiltonian associated with this perturbation may be written

$$\mathcal{H}_{ED} = \hbar\left\{g_{ext}\,\sigma^+ + g_{ext}\,\sigma^-\right\}\left\{i\,a_{ext} - i\,a^\dagger_{ext}\right\} \qquad \text{[V-30]}$$

Since $\vec{\mathbf{a}}^\dagger = \vec{\mu}^\dagger \; \overleftrightarrow{\mathbf{U}}$ and $\vec{\mathbf{a}} = \overleftrightarrow{\mathbf{U}}^{-1} \; \vec{\mu}$ we see that

$$\sigma^+ = \sum_t \mu^\dagger_t \, U_{t1} \quad \text{and} \quad \sigma^- = \sum_t U^{-1}_{1t}\,\mu_t = \sum_t U_{t1}\,\mu_t \qquad \text{[V-31]}$$

and from Equation [V-6] we may write

$$\begin{aligned}\vec{\mathcal{D}} &= \left\langle a\right|\vec{\mathcal{D}}\left|b\right\rangle \sigma^+ + \left\langle b\right|\vec{\mathcal{D}}\left|a\right\rangle \sigma^- \\ &= \sum_t \left\{\left\langle a\right|\vec{\mathcal{D}}\left|b\right\rangle U_{t1}\,\mu^\dagger_t + \left\langle b\right|\vec{\mathcal{D}}\left|a\right\rangle U_{t1}\,\mu_t\right\}\end{aligned} \qquad \text{[V-32]}$$

In light of Equation [V-29], the ***standardized form*** for the frequency dependent susceptibility (see Equation [VA-12] becomes

$$\tilde{\chi}(\omega) = \frac{N\ e^2}{\hbar\,\varepsilon_0}\left|\langle a|\vec{\mathcal{D}}\,|b\rangle\right|^2 \left\{ \frac{\prod_q(\omega_q-\omega)}{\prod_s(\lambda_s-\omega)} + \frac{\prod_q(\omega_q+\omega)}{\prod_s(\lambda_s+\omega)} \right\} \qquad [\text{ V-33 }]$$

By eliminating the U's from Equations [V-24a] and [V-24b] we see that

$$\lambda_s-\omega_0-\sum_t |\,g_t\,|^2\left(\lambda_s-\omega_t\right)^{-1}=0 \qquad [\text{ V-34 }]$$

Again

$$\left[\lambda_s-\omega_0-\sum_t |\,g_t\,|^2\left(\lambda_s-\omega_t\right)^{-1}\right]\prod_q\left(\lambda_s-\omega_q\right)=0 \qquad [\text{ V-35 }]$$

so that we can write

$$\left[\omega-\omega_0-\sum_t |g_t|^2\left(\lambda_s-\omega_t\right)^{-1}\right]\prod_q\left(\omega-\omega_q\right)=\prod_s\left(\omega-\lambda_s\right) \qquad [\text{ V-36 }]$$

Therefore

$$\tilde{\chi}(\omega) = \frac{N\ e^2}{\hbar\,\varepsilon_0}\left|\langle a|\vec{\mathcal{D}}\,|b\rangle\right|^2 \left\{ \frac{1}{\omega-\omega_0-\sum_t |\,g_{\mathrm{t}}\,|^2\left(\omega_t-\omega\right)^{-1}} + \frac{1}{\omega+\omega_0-\sum_t |\,g_{\mathrm{t}}\,|^2\left(\omega_t+\omega\right)^{-1}} \right\} \qquad [\text{ V-37 }]$$

where the sum $\sum_t |\,g_{\mathrm{t}}\,|^2\left(\omega_t-\omega\right)^{-1}$ gives an explicit, non-phenomenological accounting of interactions with the cavity modes and hence of spontaneous emission!

EVALUATION OF SPONTANEOUS EMISSION RATE:

Recall the discussion of phenomenologically defined damping in semiclassical models of the dielectric response function in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*. Recollect, in particular, Equation [III-19c] in those notes. In reconciling that discussion with the content of Equation [V-37] above, we see that the summation $\sum_t |g_t|^2 (\omega_t - \omega)^{-1}$ replaces the simple damping parameter γ. Our task here is evaluate this integral which we write as

$$i\,\tilde{\gamma}(\omega) = \lim_{\varepsilon\to 0}\sum_t \frac{|g_t|^2}{\omega_t - \omega - i\varepsilon} = \lim_{\varepsilon\to 0}\int \frac{|g_t|^2}{\omega_t - \omega - i\varepsilon}\,\rho_{\omega_t}(\omega_t)\,d\omega_t\,. \qquad [\text{ V-38 }]$$

where $\rho_{\omega_t}(\omega_t)\,d\omega_t$ is number of cavity modes with frequency between ω_t and $\omega_t + d\omega_t$.. Following arguments best explicated long ago by Heitler,[30] it may be shown that

---

[30] W. Heitler, in Chapter II, Section 8 of *The Quantum Theory of Radiation* (3rd edition), Oxford Press (1954) uses contour integral arguments to shown that

$$\lim_{\varepsilon\to 0}\frac{1}{x - i\varepsilon} = \mathcal{P}r\;\frac{1}{x} + i\,\pi\,\delta(x)\,.$$

To quote W. H. Louisell's summary in Chapter 5 of *Radiation and Noise in Quantum Electronics*,

> "…if $f(x)$ is well behaved and has no poles at $x = a$, then
>
> $$\int_{-\infty}^{\infty}\frac{f(x)}{x-a}\,dx = \mathcal{P}r\int_{-\infty}^{\infty}\frac{f(x)}{x-a}\,dx + i\,\pi f(x)$$
>
> where $\mathcal{P}r$ means the Cauchy principle part and is defined by
>
> $$\mathcal{P}r\int_{-\infty}^{\infty}\frac{f(x)}{x-a}\,dx = \lim_{\mu\to 0}\left[\int_{-\infty}^{a-\mu}\frac{f(x)}{x-a}\,dx + \int_{a+\mu}^{\infty}\frac{f(x)}{x-a}\,dx\right]$$
>
> provided the limit on the right side exists."

$$i\,\tilde{\gamma}(\omega) = \mathcal{P}r\int_0^\infty \frac{|\,g_t\,|^2}{\omega_t-\omega}\,\rho_{\omega_t}(\omega_t)\;d\omega_t + i\,\pi\int |\,g_t\,|^2\rho_{\omega_t}(\omega_t)\,\delta(\omega_t-\omega)\;d\omega_t\,..\quad [\text{ V-39 }]$$

which we write as $i\,\tilde{\gamma}(\omega) = \Delta(\omega) + i\,\gamma(\omega)$. **This is an extremely important result!!** It shows that the interaction between the atom and the cavity modes leads to a frequency shift or correction in the atomic splitting ω_{ab}

$$\Delta(\omega) = \mathcal{P}r\int_0^\infty \frac{|\,g_t\,|^2}{\omega_t-\omega}\,\rho_{\omega_t}(\omega_t)\;d\omega_t \qquad [\text{ V-40a }]$$

and a spontaneous emission decay rate

$$\gamma(\omega) = \pi|\,g_t\,|^2\,\rho_{\omega_t}(\omega)\ . \qquad [\text{ V-40b }]$$

If we assume that the cavity modes defined for the blackbody calculation in Section IV of the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory* (see Equation [IV-5] in those notes) are the appropriate modes, we know that

$$\rho_{\omega_t}(\omega) = \omega^2/c^3\ \pi^2$$

and from Equation [V-9] we know that

$$|\,g_t\,|^2 = \frac{\omega_t\,e^2}{2\,\hbar\,\varepsilon_0}\left|\langle a|\vec{\mathcal{D}}|b\rangle\ \hat{\mathbf{e}}_t\right|^2\ .$$

Treating the shift $\Delta(\omega)$, the radiative correction to atomic energy level separation, is a very complex and much studied matter. The simple interpretation of Equation [V-38a] is problematic since the integrand is proportional to ω_t^2 at large ω_t and, thus, the correction significantly diverges!!

The divergence in $\Delta(\omega)$ was for many years an unresolved discrepancy between the quantum theory of radiation and observational spectroscopy. The difficulty was overcome by Bethe in 1947[31] using a technique known as mass renormalization. Bethe point out that the divergence can mainly be associated with the mass of the electron. It is found that the energy of a free electron has an infinite contribution arising from the interaction of the electron with the electromagnetic field. In other words, the apparent mass of the electron is shifted by an infinite amount from the mass of an electron which is not in interaction with the radiation field. However, the former mass is the one measured experimentally, since it is never possible to isolate an electron from the radiation field. Identification of the measured electron mass with the theoretical mass, after renormalization to take account of the energy of interaction with the radiation field, removes most of the divergence from $\Delta(\omega)$……

Calculations for the hydrogen atom show that $\Delta(\omega)$ vanish unless one of the states in the transition is an S state. Even when it does not vanish, the renormalized $\Delta(\omega)$ is always very small compared with the excitation frequency ω_0, and varies slowly with ω. For example, the magnitude of $\Delta(\omega)$ for the $2\,^2S_{1/2}$ state of hydrogen is about 10^9 Hz, or roughly six orders of magnitude smaller than the $n = 2$ state excitation energy..… The existence of level shifts was first demonstrated by Lamb and Retherford in experiments on radiative transition between the $2\,^2S_{1/2}$ state of hydrogen and the unshifted $2\,^2P_{1/2}$ state. The splitting between these states is known as the Lamb shift. [32]

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[31] Bethe, H. A., *Phys. Rev*. **72**, 339 (1947)

[32] From Chapter 8, Rodney Loudon, *Quantum Theory of Light* (1st edition), Oxford (1973)

However Equation [V-38b] is not complicated by divergences and, consequently, we easily obtain the famous Weisskopf-Wigner formul[33] for the spontaneous emission decay rate into the modes of a three-dimensional cavity

$$\gamma(\omega) = \pi \left|g_t\right|^2 \rho_{\omega_t}(\omega) = \frac{e^2\,\omega^3}{2\pi\varepsilon_0\,\hbar c^3} \left|\langle a|\vec{\mathcal{D}}\,|b\rangle\ \hat{\mathbf{e}}_t\right|^2$$

$$\frac{1}{4\,\pi\varepsilon_0}\,\frac{2\omega^3}{3\,\hbar c^3}\left|\ \right|^2 \quad . \qquad [\text{ V-41 }]$$

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[33] V. Weisskopf and E. Wigner, Z. Phys., **63**, 54 (1930).

APPENDIX: THE DIELECTRIC SUSCEPTIBILITY
A General *Dressed State* Formulation

Suppose that the complete Hamiltonian of a coupled system is parsed into two components

$$\mathcal{H} = \mathcal{H}_o + \mathcal{H}_{ex} \, . \qquad \text{[VA-1]}$$

The component $\mathcal{H}_o$ includes the Hamiltonians for the unperturbed material system, the free radiation field and interactions of the material system with available *cavity modes*. The component $\mathcal{H}_{ex}$ is the Hamiltonian for the interactions which couple the material system to **externally excited modes**. As the first step in finding a fully quantal expression for the dielectric susceptibility, let us expand the state vector in the Schrödinger picture in terms of, presumably, known eigenkets of $\mathcal{H}_o$ -- *viz*. the ***dressed states*** of the unperturbed system --

$$\left|\psi(t)\right\rangle = \sum_s C_s(t) \exp\left(-i \, \lambda_s \, t\right) \left|s\right\rangle \, . \qquad \text{[VA-2]}$$

Following a now familiar track, we can use the Schrödinger equation of motion -- *i.e.*

$$i \hbar \frac{d}{dt}\left|\psi(t)\right\rangle = \left[\mathcal{H}_o + \mathcal{H}_{ex}\right] \left|\psi(t)\right\rangle \qquad \text{[VA-3]}$$

to obtain

$$\begin{aligned} i \, \hbar \dot{C}_r(t) &= \sum_q C_q(t) \, \exp\left[-i \left(\lambda_q - \lambda_r\right) t\right] \, \left\langle r\right|\mathcal{H}_{ex}\left|q\right\rangle \\ -i \, \hbar \dot{C}_r^{*}(t) &= \sum_q C_q^{*}(t) \, \exp\left[+i \left(\lambda_q - \lambda_r\right) t\right] \, \left\langle q\right|\mathcal{H}_{ex}\left|r\right\rangle \end{aligned} \, . \qquad \text{[VA-4]}$$

In turn, we obtain the following expansion for the time dependent expectation value of induced material system dipole moment:

$$\begin{aligned}
\left\langle\, \vec{\mathbf{p}}(t)\, \right\rangle &= -\langle \psi(t) |\, e\,\vec{\mathcal{D}}\, | \psi(t) \rangle \\
&= -\sum_r C_r(t)\, \exp\left[\, i\,\lambda_r\, t\,\right]\, \langle r |\, e\,\vec{\mathcal{D}}\, \sum_s C_s(t)\, \exp\left[-i\,\lambda_s\, t\right]\, | s \rangle \qquad \text{[VA-5]} \\
&= -\sum_r \sum_s C_r(t) C_s(t)\, \exp\left[\, i\left(\lambda_r - \lambda_s\right) t\,\right]\, e\,\langle r |\, \vec{\mathcal{D}}\, | s \rangle
\end{aligned}$$

Differentiating this expression with respect to time and using Equation [VA-4] we obtain

$$\begin{aligned}
\left\langle\, \dot{\vec{\mathbf{p}}}(t)\, \right\rangle = &-\sum_r \sum_s \left\{ i \sum_q C_q(t)\, C_s(t)\, \exp\left[\, i\left(\lambda_q - \lambda_s\right) t\,\right]\, \langle q | \mathcal{H}_{ex} | r \rangle \right\} \frac{e}{\hbar} \langle r |\, \vec{\mathcal{D}}\, | s \rangle \\
&+ \sum_r \sum_s \left\{ i \sum_q C_r(t)\, C_q(t)\, \exp\left[-i\left(\lambda_q - \lambda_r\right) t\right]\, \langle s | \mathcal{H}_{ex} | q \rangle \right\} \frac{e}{\hbar} \langle r |\, \vec{\mathcal{D}}\, | s \rangle \qquad \text{[VA-6a]} \\
&- \sum_r \sum_s C_r(t) C_s(t)\;\; i\left(\lambda_r - \lambda_s\right) \exp\left[\, i\left(\lambda_r - \lambda_s\right) t\,\right]\;\; e\langle r |\, \vec{\mathcal{D}}\, | s \rangle
\end{aligned}$$

Regrouping, we see that this expression can be written

$$\begin{aligned}
\left\langle \dot{\vec{\mathbf{p}}}(t) \right\rangle = &-\sum_r \sum_s \left\{ \frac{i\,e}{\hbar} \sum_q C_q(t)\, C_s(t)\, \exp\left[\, i\left(\lambda_q - \lambda_s\right) t\,\right]\, \left[\langle q | \mathcal{H}_{ex} | r \rangle \langle r |\, \vec{\mathcal{D}}\, | s \rangle - \langle r | \mathcal{H}_{ex} | s \rangle \langle q |\, \vec{\mathcal{D}}\, | r \rangle \right] \right\} \qquad \text{[VA-6b]} \\
&- \sum_r \sum_s C_r(t) C_s(t)\;\; i\left(\lambda_r - \lambda_s\right) \exp\left[\, i\left(\lambda_r - \lambda_s\right) t\,\right]\;\; e\langle r |\, \vec{\mathcal{D}}\, | s \rangle
\end{aligned}$$

Since $\mathcal{H}_{ex}$ is proportional to $\vec{\mathcal{D}}$, we see that, ***like magic***, the first term vanishes!!!

Hence,

$$\left\langle\, \dot{\vec{\mathbf{p}}}(t)\, \right\rangle = -\sum_r \sum_s C_r(t) C_s(t)\;\; i\left(\lambda_r - \lambda_s\right) \exp\left[i\left(\lambda_r - \lambda_s\right) t\right]\;\; e\langle r |\, \vec{\mathcal{D}}\, | s \rangle\ . \qquad \text{[VA-6c]}$$

Differentiating this expression with respect to time and, again, using Equation [VA-4] we obtain

$$\begin{aligned}\left\langle\, \ddot{\vec{\mathbf{p}}}(t)\,\right\rangle = -\sum_r\sum_s\sum_q C_q(t)C_s(t)\,\frac{e}{\hbar}\,\exp\!\left[\,i\left(\lambda_q-\lambda_s\right)\,t\right] \\ \times\left[\left(\lambda_r-\lambda_s\right)\langle q|\mathcal{H}_{ex}|r\rangle\langle r|\vec{\mathcal{D}}\,|s\rangle + \left(\lambda_r-\lambda_q\right)\langle q|\vec{\mathcal{D}}\,|r\rangle\langle r|\mathcal{H}_{ex}|s\rangle\right]\,. \\ -\sum_r\sum_s C_r(t)C_s(t)\left(\lambda_r-\lambda_s\right)^2\exp\!\left[\,i\left(\lambda_r-\lambda_s\right)\,t\right]\;e\langle r|\vec{\mathcal{D}}\,|s\rangle\end{aligned} \qquad \text{[VA-7]}$$

Our task is to now to attempt an interpretation this very nasty expression. To that end, we make use of Equation [V-33] to write Equation [VA-6c] as

$$\begin{aligned}\left\langle\, \vec{\mathbf{p}}(t)\,\right\rangle = -\sum_r\sum_s C_r(t)C_s(t)\,\exp\!\left[\,i\left(\lambda_r-\lambda_s\right)\,t\right] \\ \times e\,\langle r|\sum_t\left\{\langle a|\vec{\mathcal{D}}\,|b\rangle\,\mathrm{U}_{t1}\,\mu_t^\dagger + \langle b|\vec{\mathcal{D}}\,|a\rangle\,\mathrm{U}_{t1}\,\mu_t\right\}|s\rangle\end{aligned}\quad . \qquad \text{[VA-8a]}$$

Using the properties of the $\mu_t^\dagger$ and μ_t operators (*viz.* $|s\rangle=\mu_s^\dagger|\mathcal{g}\rangle$ and $|\mathcal{g}\rangle=\mu_s|s\rangle$), this expression reduces to

$$\left\langle\, \vec{\mathbf{p}}(t)\,\right\rangle = -\sum_r\sum_s C_r(t)C_s(t)\,\exp\!\left[\,i\left(\lambda_r-\lambda_s\right)\,t\right]\;e\left\{\langle a|\vec{\mathcal{D}}\,|b\rangle\,\mathrm{U}_{r1}\,\delta_{s\,\mathcal{g}} + \langle b|\vec{\mathcal{D}}\,|a\rangle\,\mathrm{U}_{s1}\,\delta_{r\,\mathcal{g}}\right\} \qquad \text{[VA-8b]}$$

which may interpreted as a sum of a series of dipole moment components -- *viz.*

$$\left\langle\, \vec{\mathbf{p}}(t)\,\right\rangle = \sum_s\,\vec{\mathbf{p}}_s(t) = -\sum_s\left\{C_s(t)C_{\mathcal{g}}(t)\,\exp\!\left[\,i\,\lambda_s\,t\right]\;e\langle a|\vec{\mathcal{D}}\,|b\rangle\,\mathrm{U}_{s1} + c.c.\right\} \qquad \text{[VA-8c]}$$

Given this interpretation, we return to Equation [VA-7] and use Equation [V-6] to obtain

$$\begin{aligned}\left\langle\, \ddot{\vec{\mathbf{p}}}(t)\,\right\rangle = +\sum_r\sum_s\sum_q C_q(t)C_s(t)\,\frac{e^2}{\hbar}\left[2\lambda_r-\lambda_s-\lambda_q\right]\times\exp\!\left[\,i\left(\lambda_q-\lambda_s\right)\,t\right]\;\vec{\mathbf{E}}_{\mathrm{T}}(0)\;\langle q|\vec{\mathcal{D}}\,|r\rangle\langle r|\vec{\mathcal{D}}\,|s\rangle \\ +\sum_r\sum_s C_r(t)C_s(t)\left(\lambda_r-\lambda_s\right)^2\exp\!\left[\,i\left(\lambda_r-\lambda_s\right)\,t\right]\;e\langle r|\vec{\mathcal{D}}\,|s\rangle\end{aligned} \qquad .\text{[VA-9]}$$

Again using Equation [V-33] and the properties of the $\mu_t^\dagger$ and μ_t operators, we see that

$$
\begin{aligned}
\langle q|\vec{\mathcal{D}}|r\rangle\langle r|\vec{\mathcal{D}}|s\rangle = {} & \langle q|\sum_x\left\{\langle a|\vec{\mathcal{D}}|b\rangle\,\mathrm{U}_{x1}\,\mu_x^\dagger+\langle b|\vec{\mathcal{D}}|a\rangle\,\mathrm{U}_{x1}\,\mu_x\right\}|r\rangle \\
& \times\langle r|\sum_y\left\{\langle a|\vec{\mathcal{D}}|b\rangle\,\mathrm{U}_{y1}\,\mu_y^\dagger+\langle b|\vec{\mathcal{D}}|a\rangle\,\mathrm{U}_{y1}\,\mu_y\right\}|s\rangle \\
= {} & \sum_x\sum_y\left\{\langle a|\vec{\mathcal{D}}|b\rangle^2\,\mathrm{U}_{x1}\,\mathrm{U}_{y1}\langle q|\mu_x^\dagger|r\rangle\langle r|\mu_y^\dagger|s\rangle\right. \\
& +\left|\langle a|\vec{\mathcal{D}}|b\rangle\right|^2\,\mathrm{U}_{x1}\,\mathrm{U}_{y1}\langle q|\mu_x^\dagger|r\rangle\langle r|\mu_y|s\rangle \qquad\qquad [\text{ VA-10a}] \\
& +\langle b|\vec{\mathcal{D}}|a\rangle^2\,\mathrm{U}_{x1}\,\mathrm{U}_{y1}\langle q|\mu_x|r\rangle\langle r|\mu_y|s\rangle \\
& \left.+\left|\langle b|\vec{\mathcal{D}}|a\rangle\right|^2\,\mathrm{U}_{x1}\,\mathrm{U}_{y1}\langle q|\mu_x|r\rangle\langle r|\mu_y^\dagger|s\rangle\right\}
\end{aligned}
$$

which reduces to

$$
\langle q|\vec{\mathcal{D}}|r\rangle\langle r|\vec{\mathcal{D}}|s\rangle = \langle a|\vec{\mathcal{D}}|b\rangle\langle b|\vec{\mathcal{D}}|a\rangle\,\mathrm{U}_{q1}\,\mathrm{U}_{s1}\,\delta_{r\,g}+\langle b|\vec{\mathcal{D}}|a\rangle\langle a|\vec{\mathcal{D}}|b\rangle\left|\mathrm{U}_{r1}\right|^2\delta_{q\,g}\,\delta_{s\,g}\,[\text{ VA-10b }]
$$

Substituting this expression and the expression in Equation [V-33] into Equation [VA-9] and, again, using the properties of the $\mu_t^\dagger$ and μ_t operators it relatively straightforward to obtain

$$
\begin{aligned}
\left\langle\,\ddot{\vec{\mathbf{p}}}(t)\,\right\rangle+\sum_s\lambda_s^2\,\vec{\mathbf{p}}_s(t) = {} & -\frac{e^2}{\hbar}\sum_q\sum_s\left[\lambda_s+\lambda_q\right]C_q(t)C_s(t)\,\mathrm{U}_{q1}\,\mathrm{U}_{s1} \\
& \times\exp\left[\,i\left(\lambda_q-\lambda_s\right)t\right]\vec{\mathbf{E}}_{\mathrm{T}}(0)\;\langle a|\vec{\mathcal{D}}|b\rangle\langle b|\vec{\mathcal{D}}|a\rangle\,.[\text{ VA-11a }] \\
& +\frac{e^2}{\hbar}\sum_r 2\lambda_r\left|C_g(t)\right|^2\left|\mathrm{U}_{r1}\right|^2\vec{\mathbf{E}}_{\mathrm{T}}(0)\;\langle b|\vec{\mathcal{D}}|a\rangle\langle a|\vec{\mathcal{D}}|b\rangle
\end{aligned}
$$

Thus, in a kind of ***rotating field approximation***, we get a set of driven harmonic oscillator equations of the form

$$\ddot{\vec{\mathbf{p}}}_s(t) + \lambda_s^2\,\vec{\mathbf{p}}_s(t) \approx \frac{e^2}{\hbar}\,\vec{\mathbf{E}}_{\mathrm{T}}(0)\,\left|\langle a|\,\vec{\mathcal{D}}\,|b\rangle\right|^2\; 2\lambda_s \left[\left|C_g(t)\right|^2 - \left|C_s(t)\right|^2\right]\left|\mathrm{U}_{s1}\right|^2 . \qquad \text{[VA-11b]}$$

In the Weisskopf-Wigner approximation[1] -- *i.e.* $\left|C_g(t)\right|^2 \approx 1$ -- we can easily solve these equations and sum their results to obtain a ***standardized form*** for the frequency dependent of the **dressed** dielectric susceptibility of the system

$$\begin{aligned}\tilde{\chi}(\omega) &\approx \frac{N\,e^2}{\hbar\,\varepsilon_0}\left|\langle a|\,\vec{\mathcal{D}}\,|b\rangle\right|^2 \sum_s \frac{2\lambda_s}{\lambda_s^2-\omega^2}\left|\mathrm{U}_{r1}\right|^2 \\ &= \frac{N\,e^2}{\hbar\,\varepsilon_o}\left|\langle a|\,\vec{\mathcal{D}}\,|b\rangle\right|^2 \sum_s \left|\mathrm{U}_{r1}\right|^2 \left[\frac{1}{\lambda_s-\omega}+\frac{1}{\lambda_s+\omega}\right]\end{aligned} \qquad \text{[VA-12]}$$

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[1] V. Weisskopf and E. Wigner, Z. Phys., **63**, 54 (1930).

VI. PHOTOELECTRIC DETECTION AND PHOTON COUNTING

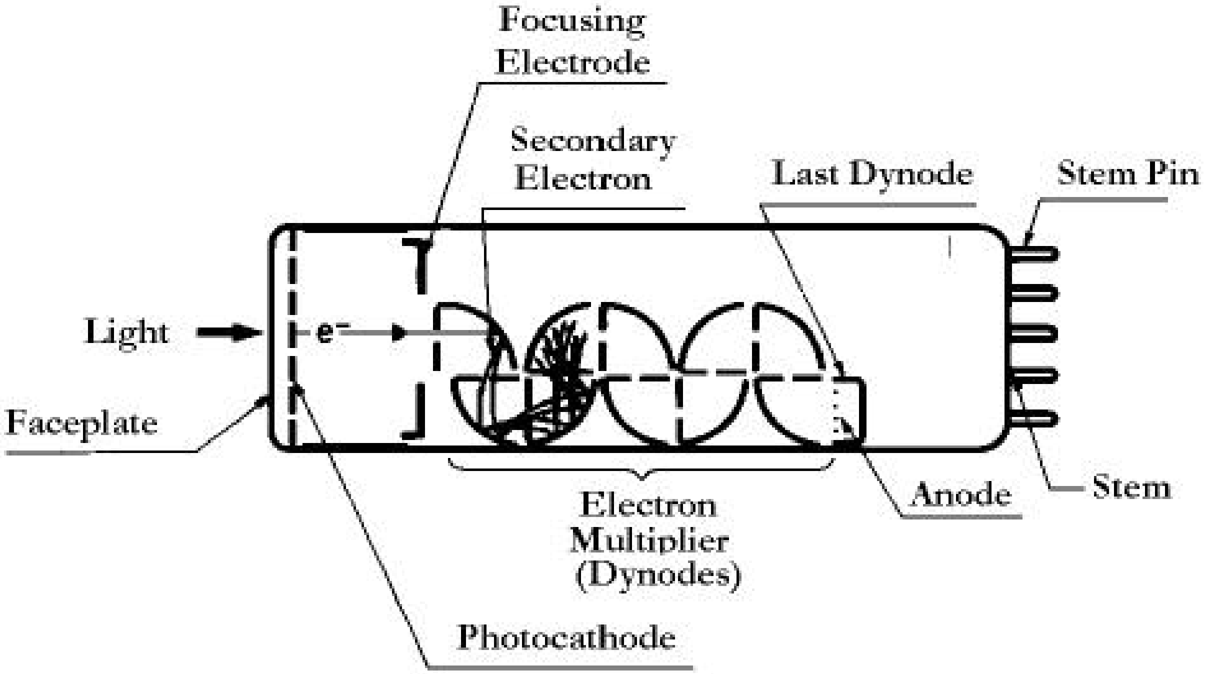
To introduce the subject let us quote Glauber in *Photon Statistics*[34]

> "Photon counters function by absorbing photons from the field. What they count are, strictly speaking, not photons but atomic photoabsorption processes rendered individuall detectable by an amplification mechanism of some sort. In most of the devices used experimental the fundamental absorption process is the photoemission of an electron. The amplifying mechanism for the photoelectrons is usually a cascade multiplier. The precise way in which amplification is achieved need not concern us here however, since it plays no direct role in determining what the counter detects.
>
> "An ideal photon counter would be one which is quite small in size and has a sensitivity independent of photon frequencies (at least over the spectral of the incident field). Both requirements can be met rather well, in fact, by the simplest possible model of a counter, a single atom which we observe to see whether and when it emits a photoelectron . Since the atom is quite small compared with the wave length of visible light most of the transitions it can undergo may be treated by means of the electric dipole approximation. in this approximation the atom is coupled to the field through the interaction Hamiltonian
>
> $$\mathcal{H} = -e \sum_{\gamma} \vec{\mathbf{q}}_{\gamma} \, \vec{\mathbf{E}}\left(\vec{\mathbf{r}}, t\right)$$
>
> in which $\vec{\mathbf{q}}_{\gamma}$ is the spatial coordinate of the γth electron of the atom relative to its nucleus which is located at $\vec{\mathbf{r}}$.

---

[34] From *Fundamental Problems in Statistical Mechanics II* (edited by E. G. D. Cohen), North-Holland Publishing (1968), pp. 140-187.

> "The intensity of a field is proportional to the rate at which a counter records photons, or in the case of our single-atom counter to the probability per unit time of our observing a photoabsorption process. …"



With careful designed, high gain photomultipliers, it is possible to register the atomic ionization caused by individual, isolated photons. Following Glauber then we will take the ***photoelectric interaction Hamiltonian*** as

$$\mathcal{H}_{PE} = -e \sum_n \vec{\mathbf{r}}_n \; \vec{\mathbf{E}}\left(\vec{\mathbf{R}},t\right) \qquad [\text{ VI-1 }]$$

where the $\vec{\mathbf{r}}_n$'s are the relative spatial coordinates of the electrons bound to a nucleus located at $\vec{\mathbf{R}}$. First-order perturbation theory informs us that the transition amplitudes associated with the process of photoelectric absorption are proportial to the matrix elements

$$\langle\, f \,|\, \mathcal{H}_{PE} |\, i \,\rangle = -e \sum_{n} \langle\, f \,|\, \vec{\mathbf{r}}_n \; \left[\vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}, t\right) + \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}, t\right) \right] |\, i \,\rangle \qquad \text{[VI-2]}$$

where *f* and *i* signify, respectively, the *final* and *initial* states of the complete electron-photon system (See, for example, Equation [II-16] in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*. Recall from Equation [II-24a] that

$$\begin{aligned} \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}, t\right) &= \sum_{\vec{\mathbf{q}}} \mathcal{E}_{\vec{\mathbf{q}}}\, \vec{\varepsilon}_{\vec{\mathbf{q}}}\; i\, a_{\vec{\mathbf{q}}}\; \exp\!\left[-i\, \omega_{\vec{\mathbf{q}}}\, t + i\; \vec{\mathbf{q}} \;\; \vec{\mathbf{R}} \right] \\ \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}, t\right) &= \sum_{\vec{\mathbf{q}}} \mathcal{E}_{\vec{\mathbf{q}}}\, \vec{\varepsilon}_{\vec{\mathbf{q}}} \left(-i\, a_{\vec{\mathbf{q}}}^{\dagger} \right) \exp\!\left[+i\, \omega_{\vec{\mathbf{q}}}\, t - i\; \vec{\mathbf{q}} \;\; \vec{\mathbf{R}} \right] \end{aligned} \qquad \text{[VI-3]}$$

If the system starts in the lowest electronic energy state and energy is conserved in the transistions, only destruction operators contribute in first-order and, hence, the rate of photoemissive transitions from one particular initial electronic state (i.e. the transistion probability of the detector atom absorbing a photon from the field at a position between times t and t+ dt) is given by

$$w_{PE}^{i}\left(\vec{\mathbf{R}}, t\right) = \sum_{f} \left| \langle\, f \,|\, \vec{\mathbf{r}} \;\; \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}, t\right) |\, i \rangle \right|^2 \qquad \text{[VI-4a]}$$

Using Equation [I-6]] in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory* and the now familiar identity $\sum_{s} |s\rangle\langle s| \equiv 1$, we see that

$$\begin{aligned} w_{PE}^{i}\left(\vec{\mathbf{R}}, t\right) &= \sum_{f} \langle\, i \,|\, \vec{\mathbf{r}} \;\; \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}, t\right) |\; f \,\rangle\langle\, f \,|\, \vec{\mathbf{r}} \;\; \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}, t\right) |\; i \,\rangle \\ &= \langle\, i \,|\, \vec{\mathbf{r}} \;\; \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}, t\right) \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}, t\right) \;\; \vec{\mathbf{r}} \,|\; i \,\rangle \end{aligned} \qquad \text{[VI-4b]}$$

If we factorize the initial state as $|\; i \rangle = |\, i = \{i', i''\}\rangle = |\; A(i')\rangle |\; F(i'')\rangle$ where $|\; A(i')\rangle$ and $|\; F(i'')\rangle$ are, respectively, the initial electronic (atomic) and field (photon) states.

$$w^{i}_{PE}\left(\vec{\mathbf{R}},t\right) = \langle A(i')|\,\vec{\mathbf{r}}^{\dagger}\ \langle F(i'')|\,\vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}},t\right)\vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}},t\right)|F(i'')\rangle\ \vec{\mathbf{r}}\,|A(i')\rangle \qquad [\text{ VI-4c }]$$

Summing over all initial states, we have the total probability of the detector atom absorbing a photon from the field at a position between times t and $t+\mathrm{d}t$.

$$\begin{aligned} w_{PE}\left(\vec{\mathbf{R}},t\right) &= \sum_{i'}\langle A(i')|\,\vec{\mathbf{r}}^{\dagger}\ \left[\sum_{i''}\langle F(i'')|\,\vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}},t\right)\vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}},t\right)|F(i'')\rangle\right]\ \vec{\mathbf{r}}\,|A(i')\rangle \\ &= \sum_{i'}\langle A(i')|\,\vec{\mathbf{r}}^{\dagger}\ \overleftrightarrow{\mathcal{G}}^{(1)}\left(\vec{\mathbf{R}},t\right)\ \vec{\mathbf{r}}\,|A(i')\rangle \qquad [\text{ VI-5 }] \end{aligned}$$

where $\overleftrightarrow{\mathcal{G}}^{(1)}\left(\vec{\mathbf{R}},t\right) = \sum_{i''}\langle F(i'')|\,\vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}},t\right)\vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}},t\right)|F(i'')\rangle w$.

Thus, the total rate of photoemission from a given atom depends on the expectation value of the operator $\overleftrightarrow{\mathcal{G}}^{(1)}\left(\vec{\mathbf{R}},t\right)$ (or $\vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}},t\right)\vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}},t\right)$) and this rate, in turn, translates -- eventually -- in the value of the observed photocurrent in the photomultiplier.

$$\left\langle\ \overleftrightarrow{\mathcal{G}}^{(1)}\left(\vec{\mathbf{R}},t\right)\right\rangle = \sum_{i''}\mathrm{Pr}\left(F(i'')\right)\langle F(i'')|\,\vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}},t\right)\vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}},t\right)|F(i'')\rangle \qquad [\text{ VI-6a }]$$

However, $\rho_{\mathrm{rad}} = \sum_{i''}\mathrm{Pr}\left(F(i'')\right)|F(i'')\rangle\langle F(i'')|$ so that we can write

$$\left\langle\ \overleftrightarrow{\mathcal{G}}^{(1)}\left(\vec{\mathbf{R}},t\right)\right\rangle = \mathrm{Tr}\left[\rho_{\mathrm{rad}}\,\vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}},t\right)\vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}},t\right)\right] \qquad [\text{ VI-6b }]$$

We may then assert that this operator is (or is proportional to) the ***photon intensity operator*** which has an expectation value proportional to the observable light intensity.[35] and is a particular value of the more general function

$$\left\langle \vec{\mathbf{G}}^{(1)}\left(\vec{\mathbf{R}}_1, t_1; \vec{\mathbf{R}}_2, t_2\right) \right\rangle = \mathrm{Tr}\left[\rho_{\mathrm{rad}}\, \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}_1, t\right) \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}_2, t_2\right)\right] \qquad [\text{ VI-7 }]$$

which is call the **first-order correlation diadic (function)**.

Hanbury Brown - Twist Effect -- Correlation of Radiation from Incoherent Sources [36]

If we extend the arguments associated with the introduction of Equation [VI-4a] to find the joint count probability of a double photexcitation

$$w_{PE}^{i}\left(\vec{\mathbf{R}}_1, t_1; \vec{\mathbf{R}}_2, t_2\right) = \sum_f \left| \left\langle f \right| \vec{\mathbf{r}}\;\; \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}_2, t_2\right) \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}_1, t_1\right)\;\; \vec{\mathbf{r}} \left| i \right\rangle \right|^2 \qquad [\text{ VI-8a }]$$

Again using Equation [I-6]] in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory* and the identity $\sum_s \left|s\right\rangle \left\langle s\right| \equiv 1$, we see that

$$\begin{aligned} w_{PE}^{i}\left(\vec{\mathbf{R}}_1, t_1; \vec{\mathbf{R}}_2, t_2\right) = {} & \left\langle A_1(i)\right| \vec{\mathbf{r}}_1^{\dagger} \left\langle A_2(i)\right| \vec{\mathbf{r}}_2^{\dagger} \mathbf{:} \left\langle F(i)\right| \\ & \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}_1, t_1\right) \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}_2, t_2\right) \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}_2, t_2\right) \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}_1, t_1\right) \left|F(i)\right\rangle \mathbf{:} \vec{\mathbf{r}}_2 \left|A_2(i)\right\rangle \vec{\mathbf{r}}_1 \left|A_1(i)\right\rangle \end{aligned} \qquad [\text{ VI-8b }]$$

---

[35] This interpretation is consistent with Equation [II-27b] which gives the Poynting vector as

$$\vec{\mathcal{S}} = \frac{c^2}{2} \sum_{\{l\}} \sum_{\sigma=1}^{2} \hbar\, \vec{\mathbf{k}}_{\{l\}} \left\{ a_{\{l\}\,\sigma}\; a_{\{l\}\,\sigma}^{\dagger} + a_{\{l\}\,\sigma}\; a_{\{l\}\,\sigma}^{\dagger} \right\} = c^2 \sum_{\{l\}} \sum_{\sigma=1}^{2} \hbar\, \vec{\mathbf{k}}_{\{l\}}\;\; \mathcal{N}$$

Therefore,

$$\left\langle I\left(\vec{\mathbf{R}}, t\right) \right\rangle = 2\varepsilon_0\, c\, \mathrm{Tr}\left[\rho_{\mathrm{rad}}\, \mathbf{E}^{(-)}\left(\vec{\mathbf{R}}, t\right) \mathbf{E}^{(+)}\left(\vec{\mathbf{R}}, t\right)\right]$$

[36] R. Hanbury Brown and R. Q. Twiss, *Proc. Roy. Soc.*, **A 242**, 300 (1957).

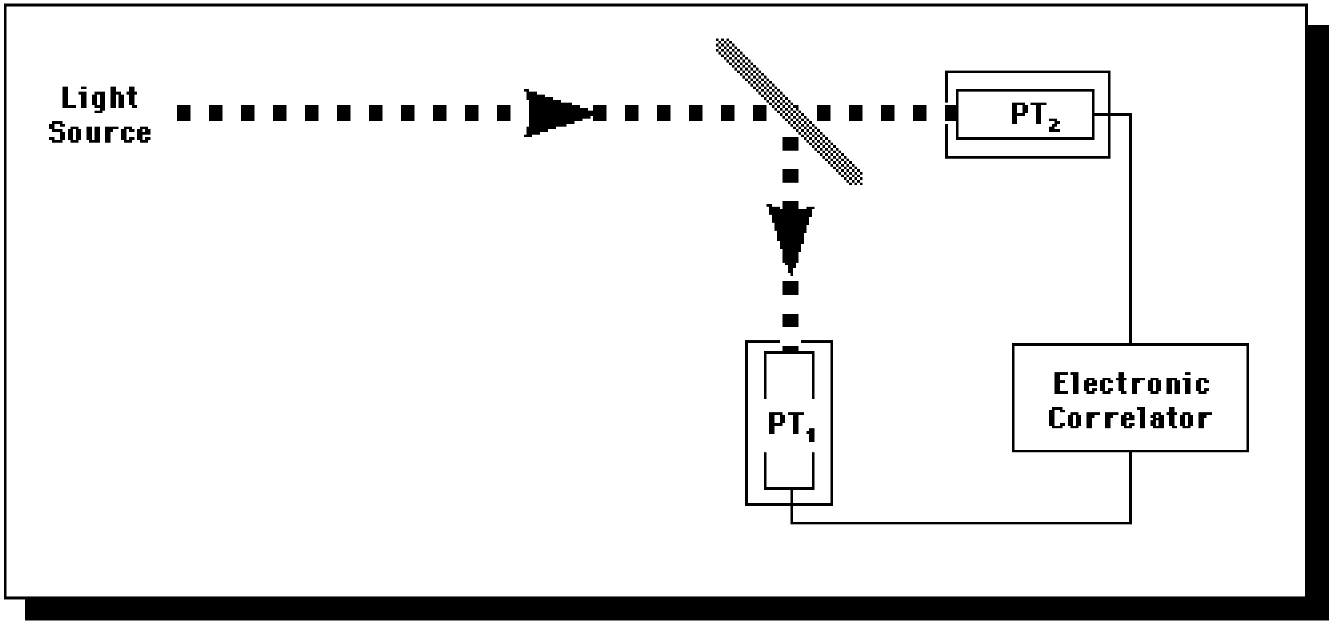
Summing over initial states

$$w_{PE}\left(\vec{\mathbf{R}}_1, t_1; \vec{\mathbf{R}}_2, t_2\right) = \sum_i \langle A_1(i)|\, \vec{\mathbf{r}}_1^{\dagger} \langle A_2(i)|\, \vec{\mathbf{r}}_2^{\dagger} : \overleftrightarrow{\mathcal{G}}^{(2)}\left(\vec{\mathbf{R}}_1, t_1; \vec{\mathbf{R}}_2, t_2\right) : \vec{\mathbf{r}}_2\, |A_2(i)\rangle\, \vec{\mathbf{r}}_1\, |A_1(i)\rangle \qquad [\text{ VI-8c }]$$

where

$$\overleftrightarrow{\mathcal{G}}^{(2)}\left(\vec{\mathbf{R}}_1, t_1; \vec{\mathbf{R}}_2, t_2\right) = \langle F(i)|\vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}_1, t_1\right) \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{R}}_2, t_2\right) \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}_2, t_2\right) \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}}_1, t_1\right) |F(i)\rangle \qquad [\text{ VI-8c }]$$

THE HANBURY BROWN - TWIST SPECTROMETER



Quite generally, we can follow Glauber an define the nth-order correlation function (tensor) as

$$\left\langle \overset{\leftrightarrow}{\overset{\vdots}{\overset{\leftrightarrow}{\mathbf{G}}}}{}^{(n)}\left(\vec{\mathbf{r}}_1, t_1; \vec{\mathbf{r}}_2, t_2; \ldots \vec{\mathbf{r}}_n, t_n; \vec{\mathbf{r}}_{n+1}, t_{n+1}; \ldots \vec{\mathbf{r}}_{2n}, t_{2n}\right) \right\rangle$$
$$= \mathrm{Tr}\left[\rho_{\mathrm{rad}}\, \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{r}}_1, t_1\right) \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{r}}_2, t_2\right) \ldots \vec{\mathbf{E}}^{(-)}\left(\vec{\mathbf{r}}_n, t_n\right)\, \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{r}}_{n+1}, t_{n+1}\right) \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{r}}_{n+2}, t_{n+2}\right) \ldots \vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{r}}_{2n}, t_{2n}\right)\right] \qquad \text{[VI-9]}$$

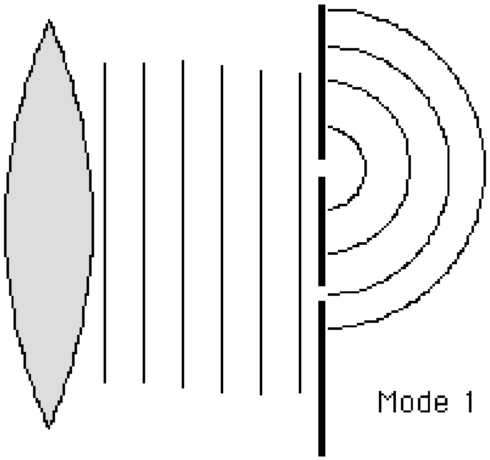
QUANTUM INTERPRETATIONS OF FUNDAMENTAL INTERFERENCE EXPERIMENTS [37]

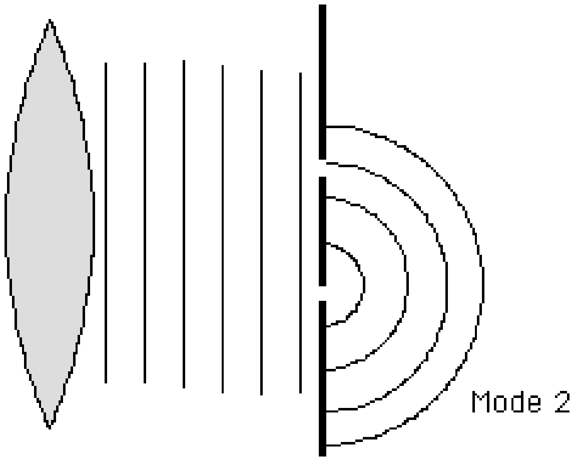
YOUNG INTERFERENCE:

To simplify things, suppose that we parse the radiation modes involved in the Young's experiment as follows:

Mode 1: Light distribution corresponding to the emergence of photons from pinhole 1. The field operators associated with this mode are $\left\{a_1, a_1^\dagger\right\}$.

Mode 2: Light distribution corresponding to the emergence of photons from pinhole 2. The field operators associated with this mode are $\left\{a_2, a_2^\dagger\right\}$.





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[37] Much of this draws heavily on excellent discussions Chapter 6 of Rodney Loudon's *The Quantum Theory of Light* (2nd edition), Oxford University Press.

Suppose that the two pinholes provide the only means of escape for the photons. The number of photons $\bar{n}_i$ which would be measured by a phototube directly behind i*th* pinhole is given by the expectation value of $a_i^{\dagger}\, a_i$ .If the pinholes are of equal size, then

$$\begin{aligned} a\ &= \left(a_1 + a_2\right)/\sqrt{2} \\ a^{\dagger} &= \left(a_1^{\dagger} + a_2^{\dagger}\right)/\sqrt{2} \end{aligned} \qquad [\text{ VI-10}]$$

(the $\left[\sqrt{2}\right]^{-1}$ factor insures that all operators satisfy the same commutation relationship). It is relative straight forward to show that

$$\begin{aligned} a_j \left[a^{\dagger}\right]^m &= \left[a^{\dagger}\right]^m a_j + \frac{m}{\sqrt{2}}\left[a^{\dagger}\right]^{m-1} \\ \left[a\right]^m a_j^{\dagger} &= a_j^{\dagger}\left[a\right]^m + \frac{m}{\sqrt{2}}\left[a\right]^{m-1} \end{aligned} \qquad [\text{ VI-11}]$$

where j = 1 or 2. Suppose that the incident state (with n photons) can be written

$$|n\rangle = \left[\sqrt{n!}\right]^{-1}\left[a^{\dagger}\right]^n |0\rangle \qquad [\text{ VI-12 }]$$

so that

$$\begin{aligned} \langle n|\, a_i^{\dagger}\, a_j\, |n\rangle &= \frac{1}{n\,!}\langle 0|\left[a\right]^n a_i^{\dagger}\, a_j \left[a^{\dagger}\right]^n|0\rangle \\ &= \frac{1}{n\,!}\langle 0|\, a_i^{\dagger}\left[a\right]^n\left[a^{\dagger}\right]^n a_j|0\rangle + \frac{1}{n!}\,\frac{n^2}{2}\langle 0|\left[a\right]^{n-1}\left[a^{\dagger}\right]^{n-1}|0\rangle \\ &= \frac{n}{2}\langle n-1|n-1\rangle = \frac{n}{2} \end{aligned} \qquad [\text{ VI-13 }]$$

Thus for a n-photon, single mode incident beam, the intensity at Q is given by

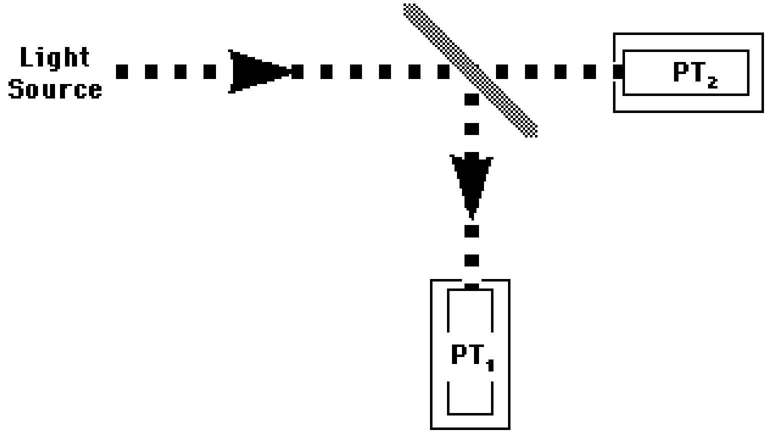
$$\begin{aligned} \langle I(Q,t)\rangle \quad & |C_1|^2\langle n|\, a_1^{\dagger}\, a_1|n\rangle + |C_2|^2\langle n|\, a_2^{\dagger}\, a_2|n\rangle \\ & +2\, C_1\ C_2\, \cos\left[k\left(s_1 - s_2\right)\right]\langle n|\, a_1^{\dagger}\, a_2|n\rangle \end{aligned} \qquad [\text{ VI-14a }]$$

or

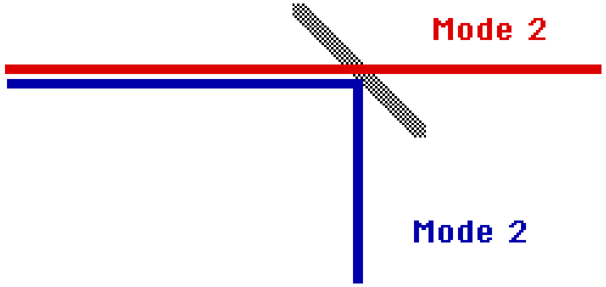
$$\langle I\,(Q\,,t)\rangle \quad \frac{n}{2}\left\{\,|C_1|^2+|C_2|^2+2\,C_1\,C_2\,\cos\left[k\left(s_1-s_2\right)\right]\,\right\} \qquad \text{[VI-14b]}$$

Thus, the same intensity distribution as observed for a large number of photons n can be built up by doing n experiments with one photon! **The interference is a one photon effect and does not depend in any way on the interaction of photons with each other.**

HANBURY BROWN- TWISS INTERFERENCE:



Experimental Configuration



Transmission and Reflection Modes

Following an argument identical to that used above, we see that the correlation between photomultiplier currents is proportional to

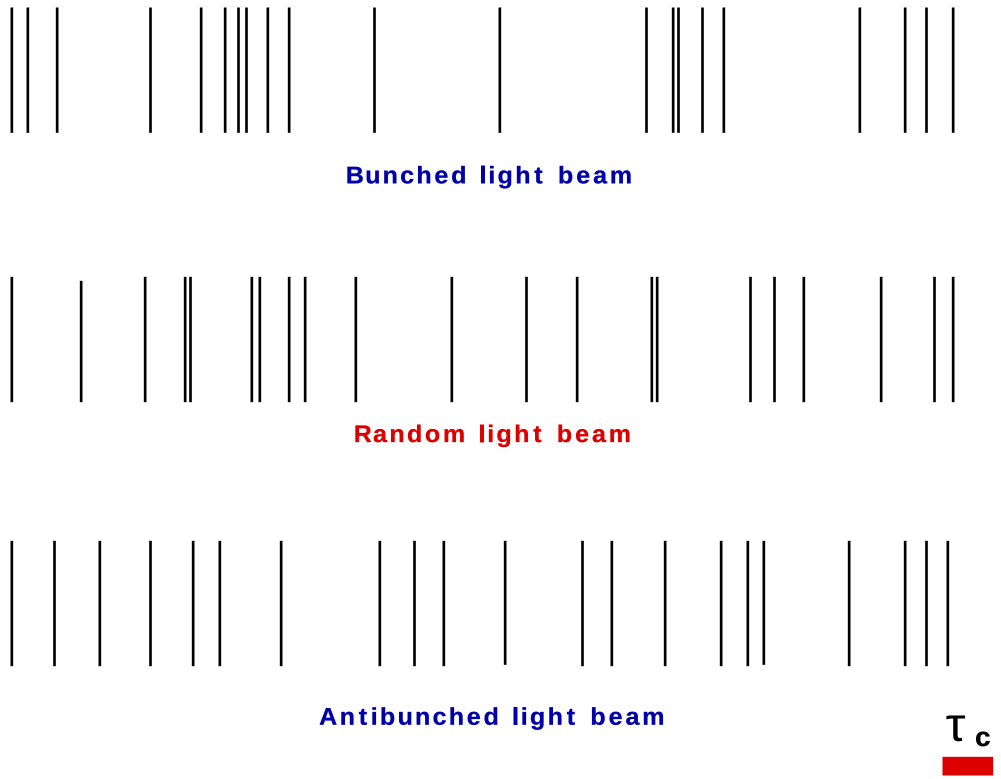
$$\left\langle a_1^{\dagger}\,a_2^{\dagger}\,a_2\,a_1\right\rangle \equiv \langle n_1\,n_2\rangle = \langle n|\,a_1^{\dagger}\,a_2^{\dagger}\,a_2\,a_1|n\rangle = \frac{n(n-1)}{4} \qquad \text{[VI-15]}$$

Thus, the degree of second-order coherence is

$$g^{(2)}(\tau) = \langle n_1\, n_2\rangle / \langle n_1\rangle\langle n_2\rangle = \frac{n(n-1)/4}{(n/2)(n/2)} = \frac{(n-1)}{n} \qquad [\text{ VI-16 }]$$

n	n_1	n_2	$\langle n_1\rangle = \langle n_2\rangle$	$\langle n_1\, n_2\rangle$	$g^{(2)}(\tau)$
1	1 0	0 1	$\frac{1}{2}$	0	0
2	2 1 1 0	0 1 1 2	1	$\frac{1}{2}$	$\frac{1}{2}$
3	3 2 2 2 1 1 1 0	0 1 1 1 2 2 2 3	$\frac{3}{2}$	$\frac{3}{2}$	$\frac{2}{3}$
4	……	……	2	3	$\frac{3}{4}$

Shematic Sequences of Photon Counts



Bunched light beam

Random light beam

Antibunched light beam

VIA. ON CLASSICAL OPTICAL INTERFERENCE, COHERENCE AND FLUCTUATIONS

Light from a real physical source is never, strictly speaking, monochromatic nor does it emanate from a single point in space. Both the amplitude and the phase of the wave field generated by a real source undergoes irregular fluctuations. Within a time interval of the order of the putative ***coherence time*** the amplitude and phase of the radiation remain relatively stable and, thus, it may be treated as monochromatic. For time intervals long compared to the coherence time, the effects of fluctuations become manifest. In particular, interference phenomena, the hallmark of wave behavior, are observed only when the interfering beams traverse paths that differ in length by an amount small compared to the ***coherence length*** -- *i.e.* the product of the coherence time and the velocity of light. In this first section we review the classical analysis and interpretation of interference effects in terms of the notions of ***partial coherence***.

COMPLEX REPRESENTATION OF POLYCHROMATIC FIELDS:

Let us first carefully define the mathematical framework of the subject. We assume that the observable optical field may be represented by a Fourier integral transform [1]

$$E^{(r)}(t) = \int_{-\infty}^{\infty} E(\omega)\ \exp[-i\,\omega t]\ d\omega \qquad \text{[VIA-1]}$$

Since of necessity $E^{(r)}(t)$ must be real

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[1] For this assumption to be valid, it is sufficient that the function $E(t)$ be ***square-integrable*** -- *i.e.*

$$\int_{-\infty}^{\infty} \left[E^{(r)}(t)\right]^2 dt < \infty \ .$$

$$E(-\omega) = E\ (\omega) \qquad \text{[VIA-2]}$$

and, consequently, Equation [VIA-1] may be re-expressed in terms of an integral over positive frequencies -- *viz*.

$$E^{(r)}(t) = \int_0^\infty \left\{ E(\omega)\, \exp[-i\,\omega t] + c.c. \right\}\, d\omega\ \cdot \qquad \text{[VIA-3]}$$

Further, if we write

$$E(\omega) = \frac{1}{2}\ a(\omega)\, \exp\big[\, i\, \varphi(\omega)\big]\ , \qquad \text{[VIA-4]}$$

where $a(\omega)$ and $\varphi(\omega)$ are both real, then the observable optical field may be expressed in the form

$$E^{(r)}(t) = \int_0^\infty a(\omega)\, \cos\left\{ i\, \left[\varphi(\omega) - \omega\, t\right]\right\} d\omega \qquad \text{[VIA-5]}$$

which is a strict generalization of the complex or ***phasor*** representation of real monochromatic fields. To complete the generalization, we follow Dennis Gabor[2] and introduce the so called complex ***analytic signal***

$$E\,(t) = \int_0^\infty a\,(\omega)\, \exp\left\{ i\, \left[\varphi(\omega) - \omega\, t\right]\right\} d\omega = 2 \int_0^\infty E\,(\omega)\, \exp[-i\,\omega t]\, d\omega \qquad \text{[VIA-6]}$$

which is to be associated with the real observable field -- *viz*. $E^{(r)}(t) = \mathbf{Re}\big[E(t)\big]$. As we shall see, this rather careful, or should we say pedantic, introduction of the analytic signal greatly facilitates and validates the use of complex quantities in handling *cycle-averaged* properties -- *e.g.* energy and intensity -- of polychromatic optical fields.[3] In most applications of interest in optics, the spectral amplitudes will have appreciable values only within a spectral width $\Delta\omega$ which is small compared to some mean frequency $\langle\omega\rangle$. Thus, it often useful to express the *analytic signal* in terms of a temporal modulation of a mean monochromatic -- *viz.*

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[2] D. Gabor, *J. Inst. Elec. Engrs.* (London), **93**, 2529 (1946).

$$E(t) = a(t)\exp\left\{ i\left[\varphi(t) - \langle\omega\rangle\, t\right]\right\} \; . \qquad \text{[VIA-7]}$$

Using Equation [VIA-6] we see that

$$a(t)\exp\left[i\varphi(t)\right] = 2\int_0^\infty E(\omega)\,\exp\left[-i\left(\omega - \langle\omega\rangle\right)t\right]\,d\omega = \int_{-\langle\omega\rangle}^\infty \tilde{E}(\omega)\,\exp\left[-i\,\omega t\right]\,d\omega \qquad \text{[VIA-8]}$$

where $\tilde{E}(\omega) = 2\,E\left(\omega + \langle\omega\rangle\right)$: by assumption, $\tilde{E}(\omega)$ will be appreciable only near $\omega = \langle\omega\rangle$. The following list of transforms are included for reference:

$$a(t) = \sqrt{\left[E^{(r)}(t)\right]^2 + \left[E^{(i)}(t)\right]^2} = \sqrt{E(t)E\,(t)} = \left|E(t)\right| \qquad \text{[VIA-9a]}$$

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[3] The following identities involving *improper functions* are invaluable when treating analytic signals -- see Beran and Parrent:

$$\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp\left(i\,x\left(y' - y\right)\right)dx = \delta\left(y' - y\right)$$

$$\frac{1}{2\pi}\int_0^\infty \exp\left[\mp\, i\,x\left(y' - y\right)\right]\,dx = \delta_\pm\left(y' - y\right) = \frac{1}{2}\left[\delta\left(y' - y\right) \mp \frac{i}{\pi}\,\frac{\mathcal{P}r}{y' - y}\right]$$

$$\varphi(t) = \langle\omega\rangle\, t + \tan^{-1}\frac{E^{(i)}(t)}{E^{(r)}(t)} = \langle\omega\rangle\, t + \tan^{-1}\left[i\,\frac{E\;(t) - E(t)}{E\;(t) - E(t)} \right] \qquad \text{[VIA-9b]}$$

$$E^{(r)}(t) = a(t)\,\cos\left\{ i\left[\varphi(t) - \langle\omega\rangle\, t\right]\right\} \qquad \text{[VIA-9c]}$$

$$E^{(i)}(t) = a(t)\,\sin\left\{ i\left[\varphi(t) - \langle\omega\rangle\, t\right]\right\}. \qquad \text{[VIA-9d]}$$

Using Equations [VIA-1] and [VIA-6], we may easily establish a form of Parseval's formula

$$\begin{aligned}
\int_{-\infty}^{\infty}\left[E^{(r)}(t)\right]^2 dt &= \int_{-\infty}^{\infty}\left[E^{(i)}(t)\right]^2 dt = \frac{1}{2}\int_{-\infty}^{\infty}\left|E\,(t)\right|^2 dt \\
&= \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} E\,(\omega)\,E\,(\omega')\,\exp\left[-i\,(\omega+\omega')\,t\right]\, d\omega\, d\omega'\, dt \qquad \text{[VIA-10]} \\
&= 2\pi\int_{-\infty}^{\infty}\left|E\,(\omega)\right|^2 d\omega = 4\pi\int_{0}^{\infty}\left|E\,(\omega)\right|^2 d\omega
\end{aligned}$$

We have thus far assumed that field is defined for all values of t. In practical terms, the field or, more likely, the observation of the field is defined only within some finite time interval $-T \le t \le T$. In all instances of any consequence, T is much larger than the physically significant times $2\pi/\Delta\omega$ and $2\pi/\langle\omega\rangle$ so that we may feel some confident in taking the idealization $T \to \infty$. However, ***stationary***[4] requires that the time averaged energy of the field, which is proportional to

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[4] A random process is characterized as ***stationary*** when all of the moments of the random variables are independent of **absolute time**.

$$\lim_{T\to\infty}\frac{1}{2T}\int_{-T}^{T}\left[E^{(r)}(t)\right]^{2}dt\,, \qquad [\text{ VIA-11 }]$$

must be finite and, thus, a stationary field function is not *square-integrable***!** The problem of analyzing such functions has been a much discussed issue in the mathematics literature.[5] However, as applied physicists, we brush these mathematical niceties under the rug in what follows. We proceed by firmly asserting that optical fields of interest do, indeed, have Fourier transforms and chance the consequences of ***trucation errors***.

The cycled-averaged intensity or power spectral density is a crucial element in the analysis of coherence effects and, from Equation [VIA-6], it is proportional

$$\begin{aligned}\left|E(\omega)\right|^{2} &= \left[\frac{1}{4\pi}\right]^{2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left[i\,\omega(t'-t)\right]E(t')E(t)\,dt\,dt' \\ &= \left[\frac{1}{4\pi}\right]^{2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left[i\,\omega\tau\right]E(t+\tau)E(t)\,dt\,d\tau\end{aligned} \qquad [\text{ VIA-12 }]$$

Consideration of this expression leads us to a logical definition of the ***first-order temporal correlation function*** of the electric field as

$$\Gamma^{(1)}(\tau) = \left\langle E(t+\tau)E(t)\right\rangle \equiv \frac{1}{2T}\int_{-T}^{T}E(t+\tau)E(t)\,dt \qquad [\text{ VIA-13 }]$$

So that Equation [VIA-12] becomes

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[5] See, in particular, N. Wiener, *Acta. Math.*,**55**, 117 (1930).

$$|E(\omega)|^2 = \left[\frac{T}{8\pi^2}\right]\int_{-\infty}^{\infty}\langle E(t+\tau)E(t)\rangle \exp[i\omega\tau]\,d\tau = \left[\frac{T}{8\pi^2}\right]\int_{-\infty}^{\infty}\Gamma(\tau)\exp[i\omega\tau]\,d\tau \qquad \text{[VIA-14]}$$

and the normalized power spectral density (distribution) is defined as

$$\begin{aligned} F(\omega) &= |E(\omega)|^2\left[\int_{-\infty}^{\infty}|E(\omega)|^2\,d\omega\right]^{-1} \\ &= \frac{1}{2\pi}\left[\int_{-\infty}^{\infty}\langle E(t+\tau)E(t)\rangle\exp[i\omega\tau]\,d\tau\right]\left[\left\langle|E(t)|^2\right\rangle\right]^{-1} \qquad \text{[VIA-15]} \\ &= \frac{1}{2\pi}\int_{-\infty}^{\infty}\gamma^{(1)}(\tau)\exp[i\omega\tau]\,d\tau \end{aligned}$$

where

$$\gamma^{(1)}(\tau) \equiv \frac{\langle E(t+\tau)\,E(t)\rangle}{\langle E(t)\,E(t)\rangle}\ . \qquad \text{[VIA-16]}$$

This normalized correlation function is usually called the field's ***complex degree of first-order temporal coherence***.[6]

YOUNG'S INTERFERENCE EXPERIMENT -- A RUDIMENTARY MEASUREMENT OF TEMPORAL COHERENCE

By considering in some detail the simplest kind of experiment in which coherence effects are important -- *i.e.* Young's (or Michelson's) observation of interference fringes -- we can

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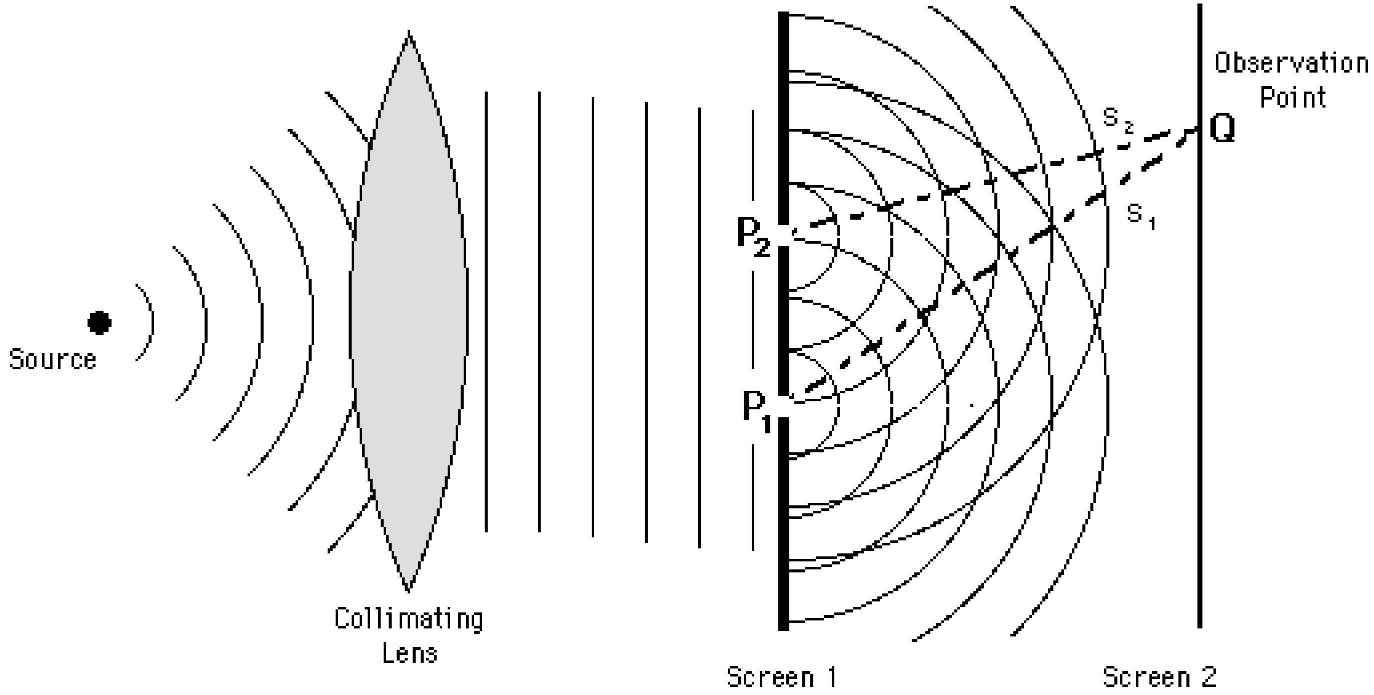
[6] It is easy to show that

$$F(\omega) = (1/\pi)\ \mathbf{Re}\int_0^{\infty}\gamma^{(1)}(\tau)\exp[i\omega\tau]\,d\tau$$

identify the significance of a light source's degree of first-order temporal coherence and demonstrate how that coherence can be measured. Consider the following basic experimental configuration:



The time integrated/averaged intensity at a given point **P** is proportional to

$$I\left(P\right) = 2\left\langle \left[E^{(r)}\left(P,t\right)\right]^2 \right\rangle = \left\langle E\left(P,t\right)\, E\ \left(P,t\right)\right\rangle \ . \qquad [\text{ VIA-17 }]$$

The real instantaneous field the point **Q** is derivable from the following analytic signal:

$$\begin{aligned} E\left(Q,t\right) &= C_1\, E\left(P_1, t - t_1\right) + C_2\, E\left(P_2, t - t_2\right) \\ &= C_1\, E\left(P_1, t - s_1/c\right) + C_2\, E\left(P_2, t - s_2/c\right) \end{aligned} \ . \qquad [\text{ VIA-18 }]$$

It follows that the time integrated/averaged intensity at the point **Q** is proportional to

$$\begin{aligned} I(Q) = {} & C_1 C_1 \langle E(P_1, t-t_1)\, E(P_1, t-t_1)\rangle + C_1 C_2 \langle E(P_1, t-t_1)\, E(P_2, t-t_2)\rangle \\ & + C_2 C_1 \langle E(P_2, t-t_2)\, E(P_1, t-t_1)\rangle + C_2 C_2 \langle E(P_2, t-t_2)\, E(P_2, t-t_2)\rangle \end{aligned} \quad . \qquad \text{[VIA-19]}$$

If the field is ***stationary***, we may with confidence shift the time origin in the various expressions so that

$$\begin{aligned} \langle E(P_n, t-t_n)\, E(P_n, t-t_n)\rangle &= \langle E(P_n, t)\, E(P_n, t)\rangle \\ &= I(P_n) = \Gamma^{(1)}_{nn}(0) \end{aligned} \qquad \text{[VIA-20a]}$$

$$\begin{aligned} \langle E(P_n, t-t_n)\, E(P_m, t-t_m)\rangle &= \langle E(P_n, t+t_m-t_n)\, E(P_m, t)\rangle \\ &= \Gamma^{(1)}_{nm}(t_m - t_n) = \Gamma^{(1)}_{nm}\left(\frac{s_m - s_n}{c}\right) . \end{aligned} \qquad \text{[VIA-20b]}$$

The normalized coherence function

$$\gamma^{(1)}_{12}(\tau) = \frac{\Gamma^{(1)}_{12}(\tau)}{\sqrt{\Gamma^{(1)}_{11}}\sqrt{\Gamma^{(1)}_{22}(0)}} = \frac{\Gamma^{(1)}_{12}(\tau)}{\sqrt{I(P_1)}\sqrt{I(P_2)}} \qquad \text{[VIA-21]}$$

is often called the ***complex mutual degree of coherence***. Finally, we write the *general interference law for stationary optical fields* as

$$I(Q) = I_1(Q) + I_2(Q) + 2\sqrt{I_1(Q)}\sqrt{I_2(Q)}\; \gamma^{(1)}_{12}\left(\frac{s_2 - s_2}{c}\right) \qquad \text{[VIA-21a]}$$

where $I_1(Q) = |C_1|^2\, I(P_1)$ and $I_2(Q) = |C_2|^2\, I(P_2)$.

Thus the degree of first-order temporal coherence may be expressed directly in terms of well-defined experimentally observable quantities -- *viz.*

$$\gamma_{12}^{(1)}\left(\frac{s_1 - s_2}{c}\right) = \frac{I(Q) - I_1(Q) - I_2(Q)}{2\sqrt{I_1(Q)}\sqrt{I_2(Q)}} \quad . \qquad \text{[VIA-21b]}$$

where $I(Q)$ and $I_n(Q)$ are, respectively, proportional to the intensity measured at Q with both pinholes open or with only the n*th* pinhole open. The ***visibility of the fringes*** given by

$$\mathcal{V}(Q) = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}} = \frac{2\sqrt{I_1(Q)}\sqrt{I_2(Q)}}{I_1(Q) + I_2(Q)}\left|\gamma_{12}^{(1)}\left(\frac{s_1 - s_2}{c}\right)\right| \qquad \text{[VIA-22]}$$

MODELS OF *CHAOTIC* LIGHT SOURCES

A COLLISION-BROADENED SOURCE -- AN EXAMPLE OF HOMOGENEOUS-BROADENING:

Perhaps, the simplest model of a *chaotic source* is that of a collision interrupted oscillator. In this model one pictures an ensemble of *atomic* radiators each of which emits a field of constant amplitude, oscillating at a fixed frequency $\langle\omega\rangle$. The phase of the continuous output of each radiator is randomly modulated by the dephasing effect of collisions which occur random at some mean collision rate of τ_0^{-1}. The figures on the next page shows samples of the field radiated per atom (left-hand graphs) and the intensity radiated per atom (right-hand graphs) for ensembles of 50, 100, 200 and 300 collision interrupt atoms.[7] . This model is treated "experimentally' in the discussion entitled *A "Toy" Model of a Randomly Fluctuating Optical Field* which is located at http://www.deas.harvard.edu/courses/ap216/lectures/ls_3/ls3_u6A/toy/toy.html. From that discussion, it is reasonable to write for a ***collision-broadened*** radiator

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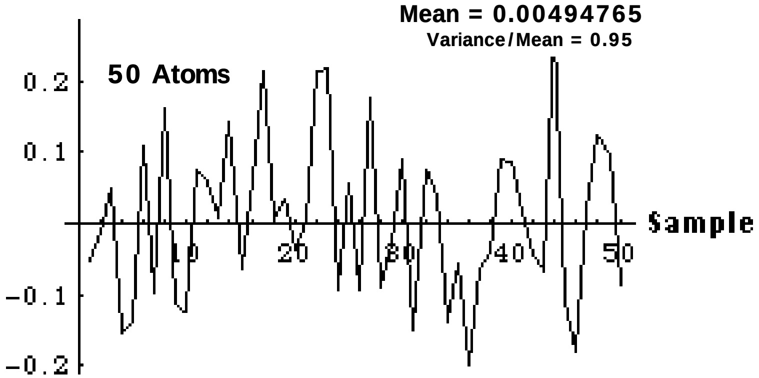
[7] The figures show samples of the sums $\left[\sum_{i=1}^{N}\sin\varphi_i\right]\Big/N$ and $\left[\sum_{i=1}^{N}\sin\varphi_i\right]^2\Big/N$ for N *atoms* with the phase radiated by each atom φ_i distributed uniformly between 0 and 2π.

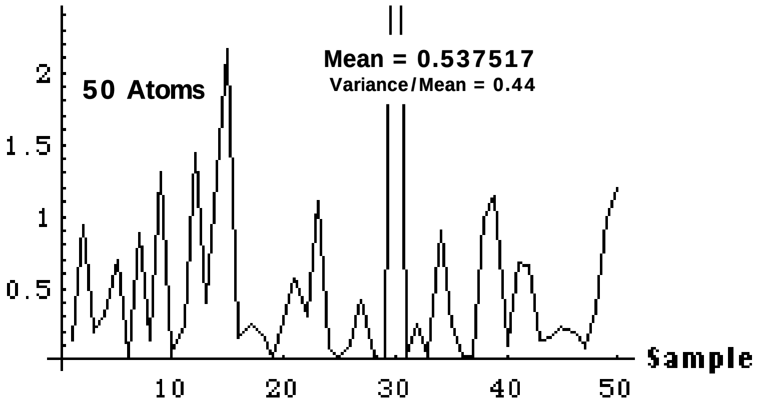
$$\gamma^{(1)}(\tau) = \exp\left[-i\langle\omega\rangle\tau - (\tau/\tau_0)\right] \qquad [\text{ VIA-23 }]$$

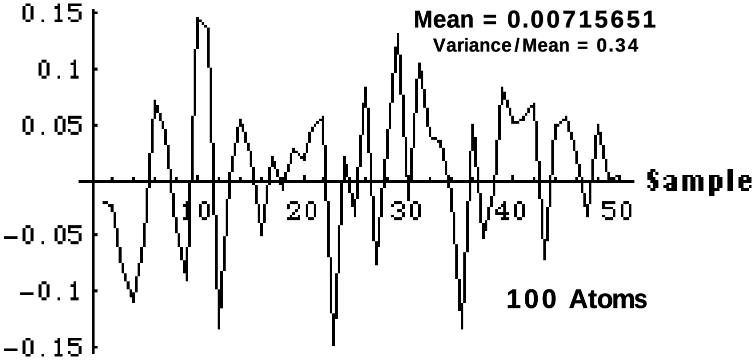
(see a graph of this function below). From Equation [VIA-15], we see that the spectrum of a collision-broadened source is given by the Lorentzian form

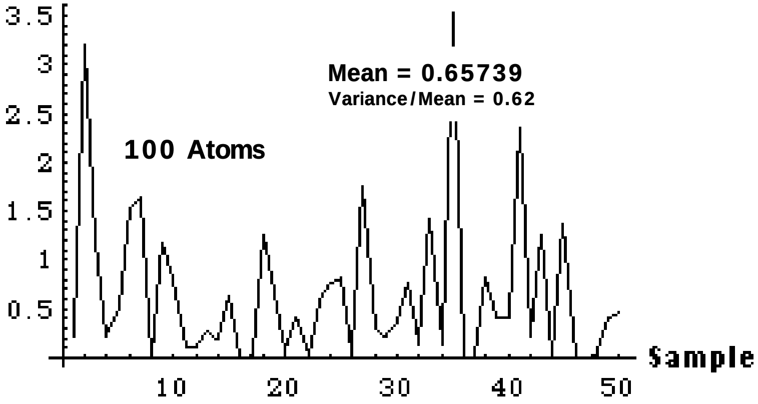
$$F(\omega) = \frac{1}{\pi\tau_0}\left[(\omega-\langle\omega\rangle)^2 + (1/\tau_0)^2\right]^{-1} \qquad [\text{ VIA-24}]$$

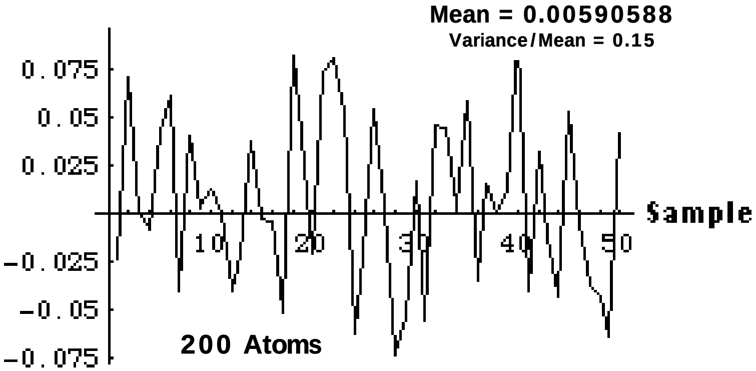
FIELD AND INTENSITY FLUCTUATIONS FOR COLLISION- OR DOPPLER-BROADENED SOURCES

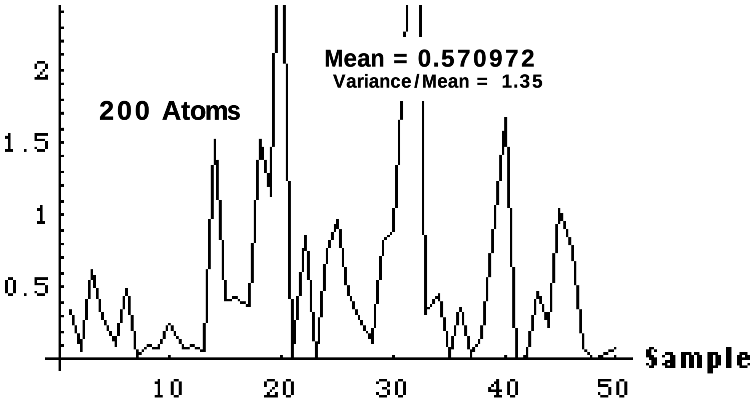





Mean = 0.00715651
Variance/Mean = 0.34
100 Atoms
Sample
0.15
0.1
0.05
-0.05
-0.1
-0.15
10
20
30
40
50


Mean = 0.65739
Variance/Mean = 0.62
100 Atoms
Sample
3.5
3
2.5
2
1.5
1
0.5
10
20
30
40
50


Mean = 0.00590588
Variance/Mean = 0.15
200 Atoms
Sample
0.075
0.05
0.025
-0.025
-0.05
-0.075
10
20
30
40
50


Mean = 0.570972
Variance/Mean = 1.35
200 Atoms
Sample
2
1.5
1
0.5
10
20
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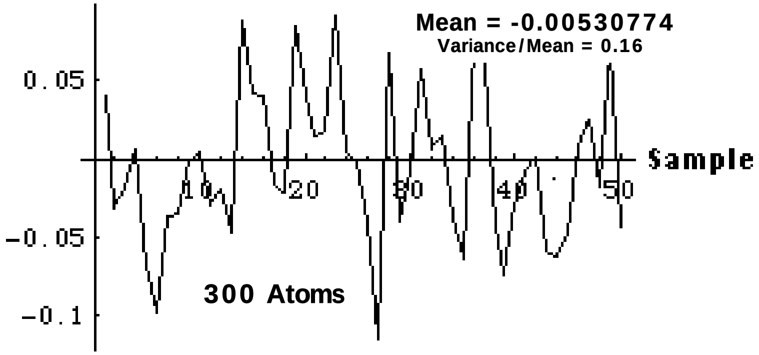

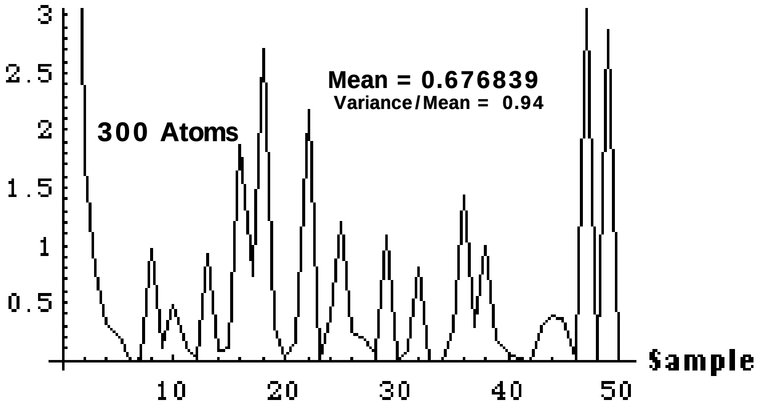

DOPPLER-BROADENED SOURCES -- AN EXAMPLE OF INHOMOGENEOUS-BROADENING:

A rather more intricate model of a *chaotic source* is that of a ***Doppler-broadening*** system of oscillators. In this model one pictures an ensemble of **randomly moving** *atomic* radiators each of which emits a field of constant amplitude, oscillating at a fixed frequency, but the observed frequency of a given atom is Doppler-shifted with respect to that fixed frequency. In the Doppler model, we write the sum of fields emitted by a ensemble of moving radiators as

$$E(t) = E_0 \sum_i \exp\left\{-i\left[\omega_i t - \varphi_i\right]\right\} \qquad [\text{ VIA-25 }]$$

where the ω_i's are the frequencies of individual atoms which are Doppler shifted from the mean radiated frequency $\langle\omega\rangle$. In this model, the effects of collisions are neglected and, consequently, the first-order correlation function is written as[8]

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[8] Since the dynamics of the individual *atoms* are uncorrelate, all cross-terms vanish.

$$\begin{aligned}\left\langle E\left(z_1 t_1\right) E\left(z_2 t_2\right)\right\rangle &= E_0^2 \sum_{i,j}\left\langle \exp\left\{ i\left[\omega_i\left(t_1 - z_1/c\right) + \varphi_i - \omega_j\left(t_2 - z_2/c\right) - \varphi_j\right]\right\}\right\rangle \\ &= E_0^2 \sum_i \left\langle \exp\left[-i\,\omega_i\,\tau\right]\right\rangle\end{aligned} \qquad \text{[VIA-26]}$$

where $\tau = t_2 - z_2/c - t_1 + z_1/c$. The sum may be converted to a integral over a Gaussian distribution of Doppler-shifted frequencies and, hence,

$$\begin{aligned}\left\langle E\left(z_1 t_1\right) E\left(z_2 t_2\right)\right\rangle &= E_0^2 \int_0^\infty \exp\left[-i\,\omega_i\,\tau\right] \exp\left\{-\left(\omega_i - \langle\omega\rangle\right)^2 \Big/ 2\delta^2\right\} d\omega_i \\ &= N_{\text{atom}}\, E_0^2 \exp\left[-i\langle\omega\rangle\,\tau - \delta^2\,\tau^2/2\right]\end{aligned} \qquad \text{[VIA-27]}$$

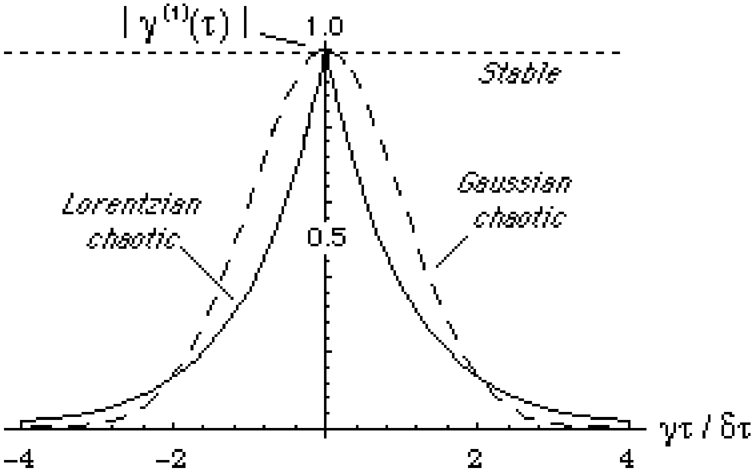
The corresponding degree of first-order temporal coherence is (see a graph of this function below)

$$\gamma^{(1)}\left(\tau\right) = \exp\left[-i\langle\omega\rangle\,\tau - \delta^2\,\tau^2/2\right] \qquad \text{[VIA-28]}$$

and the associated normalized power spectral density follows from Equation [VIA-15] as

$$F(\omega) = \left[2\pi\delta^2\right]^{-1/2} \exp\left[-\left(\omega - \langle\omega\rangle\right)^2 \Big/ 2\delta^2\right] \qquad \text{[VIA-29]}$$

DEGREE OF FIRST-ORDER COHERENCE - ABSOLUTE VALUE



SPATIAL COHERENCE: THE VAN CITTERT-ZERNIKE

In the rudimentary treatment of the Young interference experiment recapitulated above, we assume an ideal point source and, thus, neglect any effects of ***spatial coherence***. In particular, we assume that the field at points $\mathbf{P_1}$ and $\mathbf{P_2}$ due to a given radiator are in time synchronism. However, when we deal with extended sources, we must enlarge our notion of coherence The subject of spatial coherence was first developed by van Cittert and, later, it was extended by Zernike.[9] To define the problem, let us consider the following geometry:

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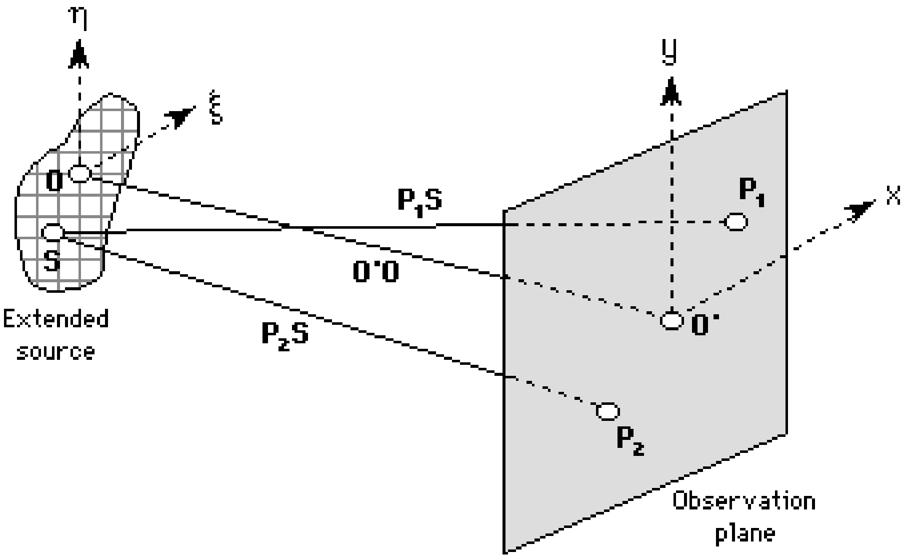
[9] See P. H. van Cittert, *Physica*, **1**, 201 (1934) and F. Zernike, *Physica*, **5**, 785 (1938).



The task before us is to determine the so called mutual intensity $J(P_1,P_2)$ due to the extended source which might be observed for the two points located on the observation plane. For simplicity, we assume that extended source is in a plane parallel to the observation plane and that it can be divided into cells of statistically independent radiators. Thus, the total field at any point on the observation plane is given by summing the fields due to each of the cells -- *viz.*

$$E(P,t) = \sum_k E_k(P,t) \quad . \qquad [\text{ VIA-30 }]$$

The mutual intensity is then defined as

$$\begin{aligned} J(P_1,P_2) &\equiv \langle E(P_1,t)\ E\ (P_2,t)\rangle \\ &= \sum_k \langle E_k(P_1,t)\ E_k(P_2,t)\rangle + \sum_{k'\neq k}\sum_k \langle E_k(P_1,t)\ E_{k'}(P_2,t)\rangle \ . \end{aligned} \qquad [\text{ VIA-31a }]$$

Because of the assumed statistical independence of the cells, the cross-terms vanish so that

$$J\,(P_1,P_2)=\sum_k \left\langle E_k(P_1,t)\;E_k(P_2,t)\right\rangle\ . \qquad \text{[VIA-31b]}$$

It the spirit of the ***Huygens principle***, we assert that component fields radiated by each coherent cell are spherical waves of the form

$$E_k\left(P,t\right)=E\left(S_k,t-\frac{P\,S_k}{c}\right)\left\{\frac{\exp\left[-i\left\langle\omega\right\rangle\left(t-\dfrac{P\,S_k}{c}\right)\right]}{P\,S_k}\right\}. \qquad \text{[VIA-32]}$$

where $E\left(S_k,t\right)$ is field radiated by each element of the source and $P\,S_k$ is, obviously, the distance between the source and observation positions. Therefore, we can write

$$\begin{aligned}\left\langle E_k\left(P_1,t\right)E_k\left(P_2,t\right)\right\rangle &=\left\langle E\left(S_k,t-\frac{P_1\,S_k}{c}\right)E\left(S_k,t-\frac{P_2\,S_k}{c}\right)\right\rangle\left\{\frac{\exp\left[\;i\left\langle\omega\right\rangle\left(P_1\,S_k-P_2\,S_k\right)/c\right]}{\left[P_1\,S_k\right]\left[P_2\,S_k\right]}\right\}\\ &=\left\langle E\left(S_k,t\right)\,E\;\left(S_k,t-\left(P_2\,S_k-P_1\,S_k\right)/c\right)\right\rangle\left\{\frac{\exp\left[\;i\left\langle\omega\right\rangle\left(P_1\,S_k-P_2\,S_k\right)/c\right]}{\left[P_1\,S_k\right]\left[P_2\,S_k\right]}\right\}\end{aligned} \qquad \text{[VIA-33]}$$

In most instances, the path difference will be small compared to the coherence length of the cell, so that, in the limit of many small cells, we obtain the famous ***van Cittert-Zernike theorem*** in the form

$$J\,(P_1,P_2)=\iint\limits_{\text{Source}} I\;(S)\,\frac{\exp\left[\;i\left\langle k\right\rangle\left(P_1\,S-P_2\,S\right)\right]}{\left[P_1\,S\right]\left[P_2\,S\right]}\,dA_S \qquad \text{[VIA-34]}$$

and the first-order degree of mutual (spatial) coherence is defined as

$$\gamma_{12} = \gamma\left(P_1, P_2\right) = \frac{J\,(P_1,P_2)}{\sqrt{I\,(P_1)}\,\sqrt{I\,(P_2)}}$$

$$= \frac{1}{\sqrt{I\,(P_1)}\,\sqrt{I\,(P_2)}} \iint\limits_{\text{Source}} I\,(S)\,\frac{\exp\left[\, i\,\langle k\rangle \left(\overline{P_1 S} - \overline{P_2 S}\right)\right]}{\left[\overline{P_1 S}\right]\left[\overline{P_2 S}\right]}\, dA_S \qquad \text{[VIA-35a]}$$

where

$$I\,(P_n) = \iint\limits_{\text{Source}} \frac{I\,(S)}{\left[\overline{P_n S}\right]^2}\, dA_S \ . \qquad \text{[VIA-35b]}$$

For most problems of interest, we may take

$$\overline{P_n S} = \sqrt{R^2 + \left(x_n - \xi\right)^2 + \left(y_n - \eta\right)^2} \approx R + \frac{\left(x_n - \xi\right)^2 + \left(y_n - \eta\right)^2}{2R} \qquad \text{[VIA-36]}$$

and, hence, we may approximate Equation [VIA-35a] as

$$\gamma_{12} \quad \frac{\displaystyle\iint\limits_{\text{Source}} I\,(\xi,\eta)\exp\left\{\, i\,\langle k\rangle \left[\frac{\left(x_1 - \xi\right)^2 - \left(x_2 - \xi\right)^2 + \left(y_1 - \eta\right)^2 - \left(y_2 - \eta\right)^2}{2R}\right]\right\} d\xi\, d\eta}{\displaystyle\iint\limits_{\text{Source}} I\,(\xi,\eta)\, d\xi\, d\eta} \qquad \text{[VIA-37a]}$$

Further, we see that in the ***Fraunhofer or far-field approximation***, the mutual coherence is the Fourier transform of the source intensity distribution -- *viz*.

$$\gamma_{12} \quad \frac{\displaystyle\iint_{\text{Source}} I(\xi,\eta)\exp\left[\,i\langle k\rangle\,\xi\,(x_1-x_2)/R\right]\,\exp\left[\,i\langle k\rangle\,\eta\,(y_1-y_2)/R\right]\,d\xi\,d\eta}{\displaystyle\iint_{\text{Source}} I(\xi,\eta)\,d\xi\,d\eta} \;\; ! \qquad \text{[VIA-37b]}$$

-- and, consequently, **the measurement of the degree of mutual coherence of a remote source provides a means to measure the angular size of that extended sources -- *i.e.* stellar objects.**

We see then that a complete first-order description of the coherence properties of a classical field is embodied in the ***complex first-order degree of spatial-temporal coherence*** which is defined as

$$\gamma^{(1)}\left(\vec{\mathbf{r}}_1 t_1, \vec{\mathbf{r}}_2 t_2\right) \equiv \frac{\left\langle E\left(\vec{\mathbf{r}}_1, t_1\right)\,E\left(\vec{\mathbf{r}}_2, t_2\right)\right\rangle}{\sqrt{\left\langle \left|E\left(\vec{\mathbf{r}}_1, t_1\right)\right|^2\right\rangle \left\langle \left|E\left(\vec{\mathbf{r}}_2, t_2\right)\right|^2\right\rangle}} \qquad \text{[VIA-38a]}$$

where

$$\left\langle E\left(\vec{\mathbf{r}}_1, t_1\right)\,E\left(\vec{\mathbf{r}}_2, t_2\right)\right\rangle \equiv \frac{1}{2T}\int_{-T}^{T} E\left(\vec{\mathbf{r}}_1, t_1 + t\right)\,E\left(\vec{\mathbf{r}}_2, t_2 + t\right)dt\,\cdot \qquad \text{[VIA-38b]}$$

FREE SPACE PROPAGATION OF COHERENCE FUNCTIONS:

We derive here an equation which allows us to generalizes the notions implicit in the van Cittert-Zernike theorem by providing us a description of how coherence propagates through space. We start by assuming that the *analytic signal* representing the field of interest satisfies an wave equation of the form

$$ {}_n^2 \, E\left(\vec{\mathbf{r}}_n, t_n\right) = \frac{1}{c^2}\frac{\partial^2}{\partial t_n^{\,2}} \, E\left(\vec{\mathbf{r}}_n, t_n\right) \qquad \text{[VIA-39]} $$

If we multiply through by $E\left(\vec{\mathbf{r}}_2, t_2\right)$, we see that

$$ \begin{aligned} {}_1^2 \, \Gamma\left(\vec{\mathbf{r}}_1 t_1, \vec{\mathbf{r}}_2 t_2\right) &= {}_1^2 \left\langle E\left(\vec{\mathbf{r}}_1, t_1\right) \, E\left(\vec{\mathbf{r}}_2, t_2\right)\right\rangle \\ &= \frac{1}{c^2}\left\langle \frac{\partial^2}{\partial t_1^{\,2}} \, E\left(\vec{\mathbf{r}}_1, t_1\right) \, E\left(\vec{\mathbf{r}}_2, t_2\right)\right\rangle . \\ &= \frac{1}{c^2}\frac{\partial^2}{\partial t_1^{\,2}} \, \Gamma\left(\vec{\mathbf{r}}_1 t_1, \vec{\mathbf{r}}_2 t_2\right) \end{aligned} \qquad \text{[VIA-40a]} $$

Similarly

$$ {}_2^2 \, \Gamma\left(\vec{\mathbf{r}}_1 t_1, \vec{\mathbf{r}}_2 t_2\right) = \frac{1}{c^2}\frac{\partial^2}{\partial t_2^{\,2}} \, \Gamma\left(\vec{\mathbf{r}}_1 t_1, \vec{\mathbf{r}}_2 t_2\right) . \qquad \text{[VIA-40b]} $$

Therefore the coherence function must satisfy the fourth-order equation

$$ {}_1^2 \; {}_2^2 \, \Gamma\left(\vec{\mathbf{r}}_1, \vec{\mathbf{r}}_2; \tau\right) = \frac{1}{c^4}\frac{\partial^2}{\partial \tau^{\,4}} \, \Gamma\left(\vec{\mathbf{r}}_1, \vec{\mathbf{r}}_2; \tau\right) \qquad \text{[VIA-41]} $$

The implication of Equations [VIA-40] and [VIA-41] is that **coherence is a field that propagates through space** and may be treat by methods which apply to other field variables. In fact, the content of the van Cittert-Zernike theorem is a description of how the coherence of extended source propagates to remote observation points. This point of view is particularly valuable in the treatment of wave propagation through random media. As a beam propagates through such a medium, the effect of random scattering events scrambles the phase of the beam and the simple notion of a field rapidly loses meaning. However, the evolution of the coherence function provides an important measure of the statistical properties of the random medium.

HIGHER-ORDER CORRELATION FUNCTIONS -- A CLASSICAL EXPLICATION OF THE FAMOUS HANBURY BROWN-TWISS EXPERIMENT:

Generalizing Equation [VIA-38a], the ***degree of nth-order spatial-temporal coherence*** can be defined as[10]

$$\gamma^{(n)}\left(\vec{\mathbf{r}}_1,t_1;\ldots\ldots;\vec{\mathbf{r}}_n,t_n;\vec{\mathbf{r}}_{n+1},t_{n+1};\ldots\ldots;\vec{\mathbf{r}}_{2n},t_{2n}\right) \equiv \frac{\left\langle E\left(\vec{\mathbf{r}}_1,t_1\right)\ldots\ldots E\left(\vec{\mathbf{r}}_n,t_n\right)E\left(\vec{\mathbf{r}}_{n+1},t_{n+1}\right)\ldots\ldots E\left(\vec{\mathbf{r}}_{2n},t_{2n}\right)\right\rangle}{\sqrt{\left\langle\left|E\left(\vec{\mathbf{r}}_1,t_1\right)\right|^2\right\rangle\ldots\ldots\left\langle\left|E\left(\vec{\mathbf{r}}_n,t_n\right)\right|^2\right\rangle\left\langle\left|E\left(\vec{\mathbf{r}}_{n+1},t_{n+1}\right)\right|^2\right\rangle\ldots\ldots\left\langle\left|E\left(\vec{\mathbf{r}}_{2n},t_{2n}\right)\right|^2\right\rangle}} \qquad [\text{ VIA-42 }]$$

In particular, the *degree of second-order temporal coherence* is defined as

$$\gamma^{(2)}\left(\vec{\mathbf{r}}_{\mathrm{ref}},\tau\right)\equiv\frac{\left\langle\left|E\left(\vec{\mathbf{r}}_{\mathrm{ref}},t\right)\right|^2\left|E\left(\vec{\mathbf{r}}_{\mathrm{ref}},t+\tau\right)\right|^2\right\rangle}{\left\langle\left|E\left(\vec{\mathbf{r}}_{\mathrm{ref}},t\right)\right|^2\right\rangle^2}=\frac{\left\langle\bar{I}\left(\vec{\mathbf{r}}_{\mathrm{ref}},t\right)\bar{I}\left(\vec{\mathbf{r}}_{\mathrm{ref}},t+\tau\right)^2\right\rangle}{\left\langle\bar{I}\left(\vec{\mathbf{r}}_{\mathrm{ref}},t\right)\right\rangle^2} \qquad [\text{ VIA-43 }]$$

We shall see that this function is an important measure of the relative timing of of intensity fluctuations.[11] As a first step, let first find an expression for the average intensity radiated by a collection of independent oscillators -- *viz*.

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[10] R. J. Glauber, in *Quantum Optics and Electronics, Les Houches, 1964* (edited by C. DeWitt, A. Blandin, and C. Cohen-Tannoudji), p63, Gordon and Breach (1965).

[11] **By the famous Schwartz inequality**

$$\left|\left\langle a^* b\right\rangle\right|^2\leq\left\langle\left|a\right|^2\right\rangle\left\langle\left|b\right|^2\right\rangle$$

so that

$$\left\langle\bar{I}\left(\vec{\mathbf{r}}_{\mathrm{ref}},t\right)\bar{I}\left(\vec{\mathbf{r}}_{\mathrm{ref}},t+\tau\right)^2\right\rangle\leq\left\langle\bar{I}\left(\vec{\mathbf{r}}_{\mathrm{ref}},t\right)^2\right\rangle\left\langle\bar{I}\left(\vec{\mathbf{r}}_{\mathrm{ref}},t+\tau\right)^2\right\rangle$$

and quite generally

$$\gamma^{(2)}\left(\vec{\mathbf{r}}_{\mathrm{ref}},\tau\right)\leq 1$$

$$\begin{aligned}\bar{I} \equiv \langle \bar{I}\,(t)\rangle = [\varepsilon_0\, c/2]\,\left\langle\, |E\,(t)|^2\right\rangle &= [\varepsilon_0\, c/2]\,\left\langle \sum_{i,j} E_i(t)\,E_j\,(t)\right\rangle \\ &= [\varepsilon_0\, c/2]\,\sum_i \left\langle\, |E_i(t)|^2\right\rangle\end{aligned} \qquad \text{[VIA-44]}$$

In this expression all of the cross-terms are presumed to vanish, since the radiation from any given atom is statistically uncorrelated with that of any other atom. For the collision- and Doppler-broadening models discussed above, the oscillators differ only in phase and this average can be expressed as

$$\bar{I} \equiv \langle \bar{I}\,(t)\rangle = N_{\text{atom}}\,[\varepsilon_0\, c/2]\,|a|^2 . \qquad \text{[VIA-45]}$$

To obtain the root-mean-square deviation in the cycle average of the intensity, we first calculate

$$\begin{aligned}\left\langle [\bar{I}\,(t)]^2\right\rangle = [\varepsilon_0\, c/2]^2\,\left\langle\, |E\,(t)|^4\right\rangle &= [\varepsilon_0\, c/2]^2\,\left\langle \sum_{i,j,k,l} E_i(t)E_j(t)E_k(t)E_l\,(t)\right\rangle \\ &= [\varepsilon_0\, c/2]^2\left\{\sum_i \left\langle\, |E_i(t)|^4\right\rangle + \sum_{i\neq j}\left\langle\, |E_i(t)|^2\,|E_j(t)|^2\right\rangle\right\} \\ &= [\varepsilon_0\, c/2]^2\left\{\sum_i \left\langle\, |E_i(t)|^4\right\rangle + \sum_{i\neq j}\left\langle\, |E_i(t)|^2\right\rangle\left\langle\, |E_j(t)|^2\right\rangle\right\} \\ &= [\varepsilon_0\, c/2]^2\left\{N_{\text{atom}}\left\langle\, |E_i(t)|^4\right\rangle + 2\,N_{\text{atom}}\,(N_{\text{atom}}-1)\left[\left\langle\, |E_i(t)|^2\right\rangle\right]^2\right\}\end{aligned} \qquad \text{[VIA-46]}$$

Again, for the collision- and Doppler-broadening models, this average can be expressed as

$$\begin{aligned}\left\langle\left[\bar{I}\,(t)\right]^2\right\rangle = \left[N_{\text{atom}}\right]^2\left[\varepsilon_0\,c/2\right]^2|a|^4\left[2-\left(N_{\text{atom}}\right)^{-1}\right]\\ 2\left[\left\langle\bar{I}\,(t)\right\rangle\right]^2\end{aligned} \qquad [\text{ VIA-47 }]$$

and, thus, we find to a very good approximation that

$$\sqrt{\left\langle\left[\bar{I}\,(t)\right]^2\right\rangle-\left[\left\langle\bar{I}\,(t)\right\rangle^2\right]}\quad \bar{I} \qquad [\text{ VIA-48 }]$$

which is consistent with the *experimental results* presented above. Let us now look at the ***two-time average of the intensity*** from a collection of statistically independent oscillators -- *viz*.

$$\begin{aligned}\left\langle\bar{I}\,(t_1)\,\bar{I}\,(t_2)\right\rangle &= \left[\varepsilon_0\,c/2\right]^2\left\langle\,\left|E\,(t_1)\right|^2\left|E\,(t_2)\right|^2\,\right\rangle\\ &= \left[\varepsilon_0\,c/2\right]^2\left\langle\,\sum_{i,j,k,l}E_i(t_1)E_j(t_2)E_k(t_1)E_l\,(t_2)\,\right\rangle\end{aligned} \qquad [\text{ VIA-49 }]$$

As a result of statistical independence, most of the cross-terms vanish so that

$$\begin{aligned}\left\langle\bar{I}\,(t_1)\,\bar{I}\,(t_2)\right\rangle = \left[\varepsilon_0\,c/2\right]^2 &\left\{\sum_i\left\langle\,\left|E_i(t_1)\right|^2\left|E_i(t_2)\right|^2\right\rangle\right.\\ &\left.+\sum_{i\neq j}\left\langle\,\left|E_i(t_1)\right|^2\left|E_j(t_2)\right|^2\right\rangle+\sum_{i\neq j}\left\langle\,E_i(t_1)\,E_i\,(t_2)\,E_j(t_1)\,E_j\,(t_2)\right\rangle\right\}\end{aligned} \qquad [\text{ VIA-50 }]$$

Again, for the collision- and Doppler-broadening models

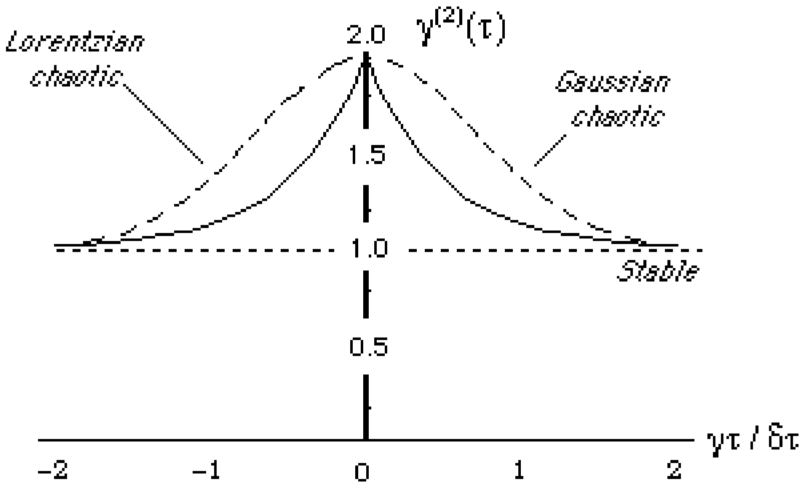
$$\left\langle \bar{I}\left(t_1\right)\bar{I}\left(t_2\right)\right\rangle = \left[\varepsilon_0\, c/2\right]^2 \Big\{ N_{\text{text}} \left\langle \left|E_i\left(t_1\right)\right|^2 \left|E_i\left(t_2\right)\right|^2 \right\rangle$$
$$+ N_{\text{text}}\left(N_{\text{text}}-1\right)\left[\left\langle \left|E_i\left(t_1\right)\right|^2 \right\rangle^2 + \left|\left\langle E_i\left(t_1\right) E_i\left(t_2\right)\right\rangle\right|^2 \right]\Big\} \qquad \text{[VIA-51]}$$

and, hence,

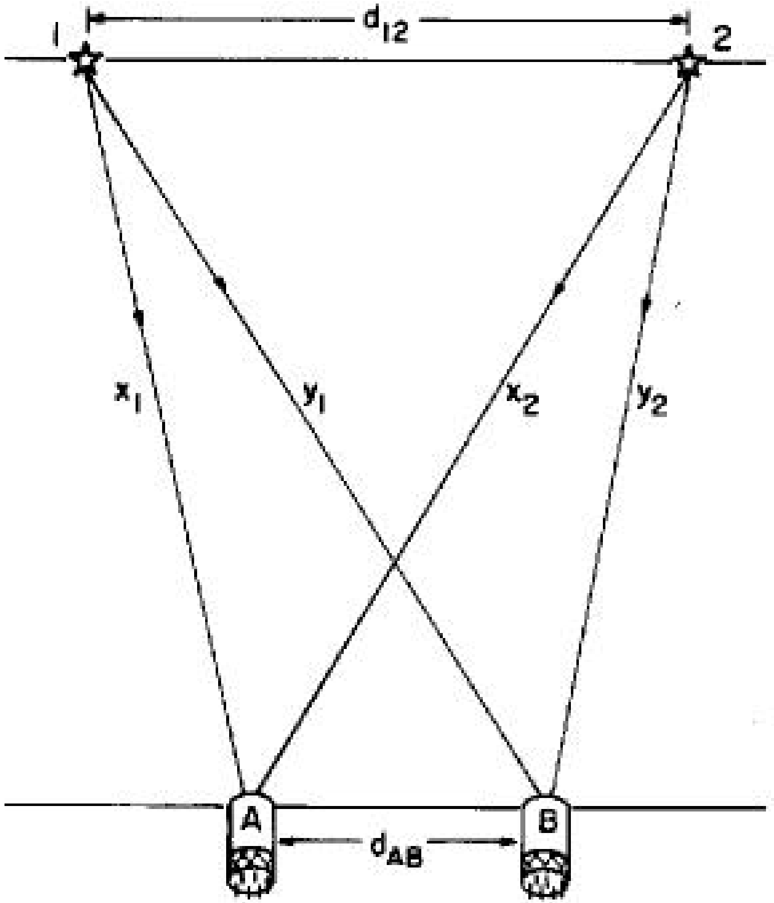
$$\gamma^{(2)}(\tau) = 1 + \left|\gamma^{(1)}(\tau)\right|^2 \qquad \text{[VIA-52]}$$

which is important result which holds for any chaotic source -- see the figure on next page. Notice that this result predicts a ***bunching*** of the intensity variations.

DEGREE OF SECOND-ORDER COHERENCE

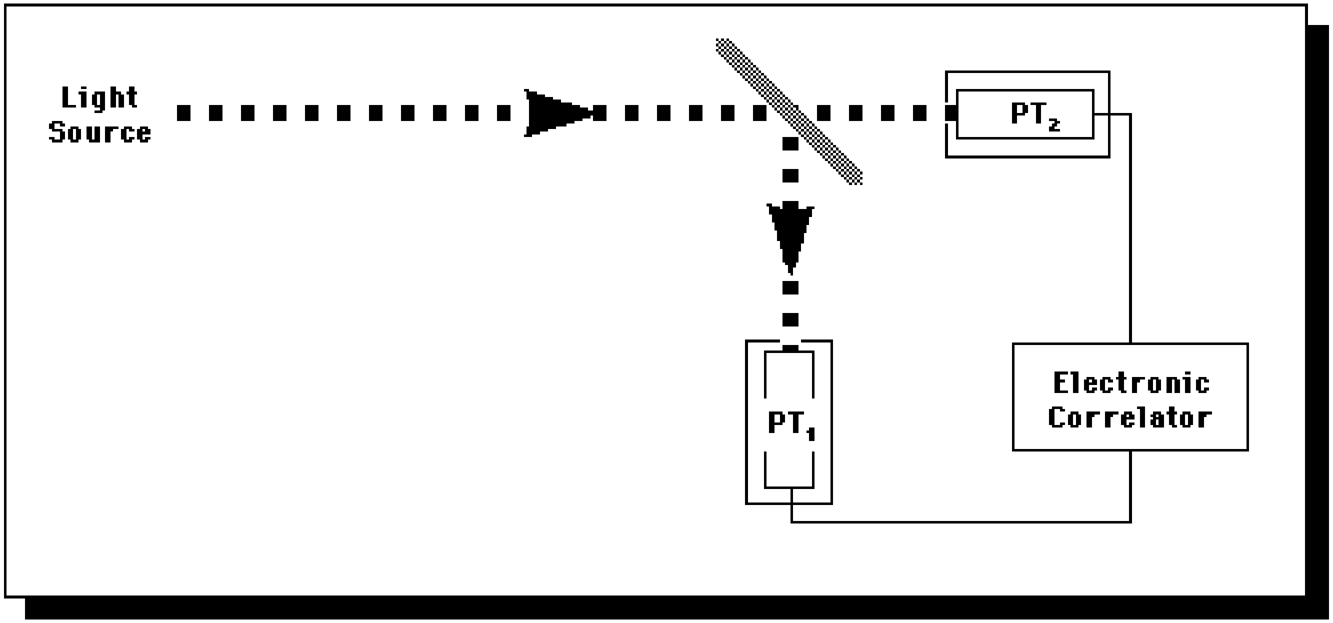


THE HANBURY BROWN - TWIST EXPERIMENT



Schematic diagram of the Hanbury Brown-Twiss experiment demonstrating coherent interference in light from incoherent sources. Sources 1 and 2 are separate by distance d12, and emit photons of identical wavelengths which are detected by detectors A and B located a distance dAB apart. The distance between the source plane and the detector plane L. A coincidence between detector outputs results in a composite signal exhibiting an interference effect depending on both d12 and dAB.

THE HANBURY BROWN - TWIST SPECTROMETER



The Hanbury Brown - Twist Spectrometer is designed to measure[12]

$$\left\langle \left[\bar{I}_1\left(\vec{\mathbf{r}}_{\mathrm{ref}}, t_1\right) - \bar{I}_1 \right] \left[\bar{I}_2\left(\vec{\mathbf{r}}_{\mathrm{ref}}, t_2\right) - \bar{I}_2 \right] \right\rangle \qquad \tfrac{1}{4}\left\{ \left\langle \bar{I}_1\left(\vec{\mathbf{r}}_{\mathrm{ref}}, t_1\right)\, \bar{I}_2\left(\vec{\mathbf{r}}_{\mathrm{ref}}, t_2\right) \right\rangle - \bar{I}^{\,2} \right\} \qquad \text{[VIA-53]}$$

This correlation may then be written

$$\frac{\left\langle \left[\bar{I}_1\left(\vec{\mathbf{r}}_{\mathrm{ref}}, t_1\right) - \bar{I}_1 \right] \left[\bar{I}_2\left(\vec{\mathbf{r}}_{\mathrm{ref}}, t_2\right) - \bar{I}_2 \right] \right\rangle}{\bar{I}_1\, \bar{I}_2} = \gamma^{(2)}(\tau) - 1 \qquad \text{[VIA-54]}$$

Generally, the results of these experiments are consistent with Equation [VIA-52], but some of the more detailed observations require a quantum mechanical interpretation.

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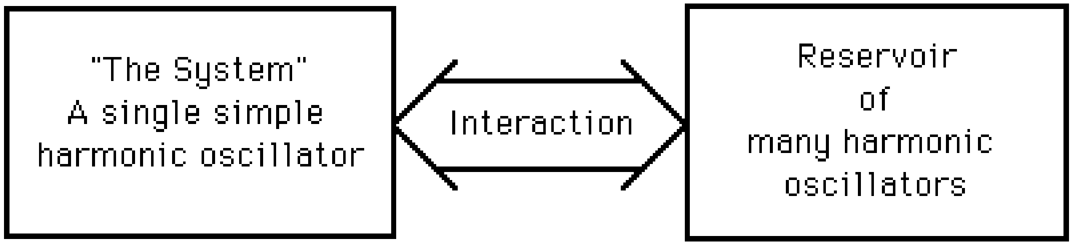
[12] R. Hanbury Brown and R. Q. Twiss, *Proc. Roy. Soc.*, **A 242**, 300 (1957).

VII. QUANTUM MECHANICAL LANGEVIN EQUATIONS [38]

A RUDIMENTARY RESEVOIR PROBLEM:[39][40]

Consider a complete system which consists of a single simple harmonic oscillator (the system component of "interest") coupled to a ***reservoir*** of many simple harmonic oscillators (the system component treated statistically)



In particular, the complete interacting system has a simple Hamiltonian of the form

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_{laser} + \mathcal{H}_{res} + \mathcal{H}_{\text{int}} \\ &= \hbar\,\Omega\, a^{\dagger} a + \sum_j \hbar\,\omega_j\, b_j^{\dagger} b_j + \sum_j \hbar\left\{ g_j\, a^{\dagger} b_j + g_j\, b_j^{\dagger} a \right\} \end{aligned} \qquad [\text{ VII-1 }]$$

where *a* is the system variable of *interest* (the "state of the system") and the b_j's are the *other* system variables (the "reservoir states"). System operators commute with reservoir operators at given time, we may easily generate the following set of equations of motion:

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[38] Much of what follows draws heavily on material in the ***Arizona Books*** -- *i.e.*

1. M. Sargent III, M. O. Scully and W. E. Lamb, Jr., *Laser Physics*, Addison-Wesley (1974)
2. P. Meystre and M. Sargent III, *Elements of Quantum Optics*, Springer-Verlag (1992)
3. Weng W. Chow, Stephan W. Koch and Murray Sargent III, *Semiconductor-Laser Physics*, Springer-Verlag (1994)

[39] This model is particular valuable in treating electromagnetic intermode coupling problems, but it also provides guidence in treating more general problems which incorporate atomic states.

[40]

$$\dot{a}(t) = i\left[\mathcal{H}, a(t)\right]/\hbar = -i\,\Omega\, a(t) - i\sum_j g_j\; b_j(t) \qquad \text{[VII-2a]}$$

$$\dot{b}_j(t) = i\left[\mathcal{H}, b_j(t)\right]/\hbar = -i\,\omega_j\; b_j(t) - i g_j\;\; a(t) \qquad \text{[VII-2b]}$$

Formally integrating Equation [VII-2b], we obtain [41]

$$b_j(t) = b_j(t_0)\; \exp\left[-i\,\omega_j\,(t-t_0)\right] - i\, g_j \int_{t_0}^{t} d\,t'\; a(t')\exp\left[-i\,\omega_j\,(t-t')\right] \qquad \text{[VII-3]}$$

The first term in this equation describes the ***free*** evolution of the reservoir states in the absence of any interaction with the system and the second gives the modification of this free evolution as a consequence of the coupling to the system. Inserting Equation [VII-3] into Equation [VII-2a] we obtain

$$\begin{aligned}\dot{a}(t) = -i\,\Omega\, a(t) - \sum_j |\, g_j\,|^2 \int_{t_0}^{t} d\,t'\, a\,(t')\exp\left[-i\,\omega_j\,(t-t')\right] \\ - i\sum_j g_j\; b_j(t_0)\exp\left[-i\,\omega_j\,(t-t_0)\right]\end{aligned} \qquad \text{[VII-4]}$$

Here the second summation describes a source of fluctuations arising from the free evolution of the reservoir states and the first is a feedback through the reservoir which might be described as a *radiation reaction*. To remove the rapid variation in the system variable we transform to the Heisenberg interaction picture -- *i.e.* we take [42]

$$a(t) = A(t)\;\exp\left[-i\,\Omega\, t\right] \qquad \text{[VII-5]}$$

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[41] As is pointed out in Section 14-3 of P. Meystre and M. Sargent III, *Elements of Quantum Optics*, Springer-Verlag (1992), the statement that "System operators commute with reservoir operators at given time…" is a very critical point. From Equation [II-3], we see that as time evolves, the reservoir operators acquire some of the characteristics of the system operator. Thus, it is crucial in this development to maintain a strict and consistent ordering of operators at different times!

[42] The transformation does not alter the system's communtation relationship, so, of necessity, $\left[A(t), A^{\dagger}(t)\right] = 1$.

so that
$$\dot{A}(t) = -\sum_j |g_j|^2 \int_{t_0}^{t} dt'\, A(t') \exp\left[-i\left(\omega_j - \Omega\right)\left(t - t'\right)\right] + F(t) \qquad [\text{ VII-6 }]$$

where
$$F(t) = -i \sum_j g_j\, b_j(t_0) \exp\left[i\left(\Omega - \omega_j\right)\left(t - t_0\right)\right] \qquad [\text{ VII-7 }]$$

is the so called ***noise operator***.

The first term on the RHS of Equation [VII-6] may be reordered as

$$\begin{aligned} &\sum_j |g_j|^2 \int_{t_0}^{t} dt'\, A(t') \exp\left[-i\left(\omega_j - \Omega\right)\left(t - t'\right)\right] \\ &\quad = \int_{t_0}^{t} dt'\, A(t') \sum_j |g_j|^2 \exp\left[-i\left(\omega_j - \Omega\right)\left(t - t'\right)\right] \end{aligned} \quad . \qquad [\text{ VII-8a }]$$

By assumption, the frequencies of the reservoir oscillators are closely spaced and widely distribution so that the function $\sum_j |g_j|^2 \exp\left[-i\left(\omega_j - \Omega\right)\left(t - t'\right)\right]$ is very sharply peaked at $t' = t$ and has a width of the order of the inverse of the reservoir's frequency bandwidth. Thus, $A(t')$ may taken out of the integration and the limits of the integration may be extended to infinite -- *i.e.*

$$\begin{aligned} &\sum_j |g_j|^2 \int_{t_0}^{t} dt'\, A(t') \exp\left[-i\left(\omega_j - \Omega\right)\left(t - t'\right)\right] \\ &\quad A(t) \sum_j |g_j|^2 \int_{-\infty}^{+\infty} dt'\, \exp\left[-i\left(\omega_j - \Omega\right)\left(t - t'\right)\right] \end{aligned} \quad . \qquad [\text{ VII-8b }]$$

Again following arguments explicated by Heitler,[43] we see that

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[43] In Chapter II, Section 8 of W. Heitler, *The Quantum Theory of Radiation* (3rd edition)

$$\int_{-\infty}^{+\infty} dt' \, \exp\left[-i\left(\Omega-\omega_j\right)\left(t-t'\right)\right] \quad \pi \;\delta\left(\Omega-\omega_j\right) - \mathcal{P}r \; \frac{i}{\Omega-\omega_j} \qquad [\text{ VII-9 }]$$

If we neglect the imaginary part of Equation [VII-9], which leads to a Lamb-type frequency shift, Equation [VII-6] leads directly to a quantum mechanical version of the classical Langevin equation [44] -- *viz*.

$$\dot{A}\left(t\right) = -\frac{\gamma}{2} A\left(t\right) + F\left(t\right) \qquad [\text{ VII-10 }]$$

where
$$\gamma = 2\pi\,\rho\left(\Omega\right) \left| g\left(\Omega\right) \right|^2 . \qquad [\text{ VII-11 }]$$

is the quantum mechanical Langevin *drift coefficient*. In optical applications the average *intensity* or *number operator* -- *i.e.* $\left\langle A^{\dagger}\left(t\right) A\left(t\right)\right\rangle$ -- is the experimental quantity of greatest interest. A useful equation of motion for that operator may obtained by making use the Langevin equation -- *i.e*. Equation [VII-10]. To that end we first write

$$\frac{d}{dt}\left\langle A^{\dagger}\left(t\right) A\left(t\right)\right\rangle = -\gamma\left\langle A^{\dagger}\left(t\right) A\left(t\right)\right\rangle + \left\langle F^{\dagger}\left(t\right) A\left(t\right)\right\rangle + \left\langle A^{\dagger}\left(t\right) F\left(t\right)\right\rangle \qquad [\text{ VII-12 }]$$

To obtain $\left\langle F^{\dagger}\left(t\right) A\left(t\right)\right\rangle$ and $\left\langle A^{\dagger}\left(t\right) F\left(t\right)\right\rangle$ we make use of the identity

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[44] In the classical Langevin theory of Browian motion, the equation of motion for drift velocity of a particle suspended in a liquid is given by

$$m\,\dot{u}\left(t\right) = -m\,\Gamma\,u\left(t\right) + F_u\left(t\right)$$

where Γ is a damping constant and $F_v\left(t\right)$ is a random, rapidly fluctuation force with zero mean value. In this classical problem the famous fluctuation-dissipation theorem is expressed as

$$\Gamma = \frac{\beta}{m} D_{uu} = \frac{\beta}{2\,m} \int_{-\infty}^{+\infty} d\tau \;\left\langle F_u\left(0\right) F_u\left(\tau\right)\right\rangle$$

where D_{uu} is the so called Fokker-Planck diffusion coefficient.

$$A(t) = A(t-\Delta t) + \int_{t-\Delta t}^{t} dt'\, \dot{A}(t') \qquad [\text{ VII-13 }]$$

so that

$$\begin{aligned} \left\langle F^{\dagger}(t)\, A(t) \right\rangle &= \left\langle F^{\dagger}(t)\, A(t-\Delta t) \right\rangle + \int_{t-\Delta t}^{t} dt' \left\langle F^{\dagger}(t)\, \dot{A}(t') \right\rangle \\ &= \left\langle F^{\dagger}(t)\, A(t-\Delta t) \right\rangle - \frac{\gamma}{2} \int_{t-\Delta t}^{t} dt' \left\langle F^{\dagger}(t)\, A(t') \right\rangle + \int_{t-\Delta t}^{t} dt' \left\langle F^{\dagger}(t)\, F(t') \right\rangle \end{aligned} \qquad [\text{ VII-14 }]$$

The first term on the RHS vanishes since *cause* cannot precede *effect*. That is, $A(t)$ cannot be correlated with a future value of the fluctuating force. Similarly, in the second term $\left\langle F^{\dagger}(t)\, A(t') \right\rangle$ vanishes except in the infinitesimal small inteval where $t' = t$. Therefore

$$\left\langle F^{\dagger}(t)\, A(t) \right\rangle = \int_{t-\Delta t}^{t} dt' \left\langle F^{\dagger}(t)\, F(t') \right\rangle . \qquad [\text{ VII-15a }]$$

If the random force is stationary in time

$$\left\langle F^{\dagger}(t)\, A(t) \right\rangle = \int_{-\Delta t}^{0} d\tau \left\langle F^{\dagger}(t)\, F(t+\tau) \right\rangle \qquad [\text{ VII-15b }]$$

and, finally, if Δt is long compared to the random force correlation time

$$\left\langle F^{\dagger}(t)\, A(t) \right\rangle = \int_{-\infty}^{0} d\tau \left\langle F^{\dagger}(t)\, F(t+\tau) \right\rangle = \frac{1}{2} \int_{-\infty}^{+\infty} d\tau \left\langle F^{\dagger}(0)\, F(\tau) \right\rangle . \qquad [\text{ VII-15c }]$$

Therefore Equation [VII-12], the equation of motion for the average number operator, becomes

$$\frac{d}{dt} \left\langle A^{\dagger}(t)\, A(t) \right\rangle = -\gamma \left\langle A^{\dagger}(t)\, A(t) \right\rangle + \int_{-\infty}^{+\infty} d\tau \left\langle F^{\dagger}(0), F(\tau) \right\rangle \qquad [\text{ VII-16 }]$$

which is one form of the famous fluctuation-dissipation theorem.

If the reservoir is in thermal equilibrium, we may directly evaluate the noise operator correlation to wit.

$$\left\langle F^{\dagger}(t)\,F(t')\right\rangle_{therm} = \sum_{j,\,k} g_j\; g_k \left\langle b_j^{\dagger}(0)\; b_k(0)\right\rangle_{therm} \exp\left[-i\left(\Omega-\omega_j\right)\,t\right]\,\exp\left[i\left(\Omega-\omega_k\right)\,t'\right] \qquad [\text{ VII-17 }]$$

Since the operators of the reservoir commute we can write

$$\begin{aligned}\left\langle b_j^{\dagger}(0)\; b_k(0)\right\rangle_{therm} &= \left\langle\, n_j \,\right\rangle_{therm}\delta_{jk}\\ &= \frac{\partial}{\partial\beta}\,\ln\left\{\sum_{n_j}\exp\left[-n_j\,\beta\,\hbar\omega_j\right]\right\} \qquad\qquad [\text{ VII-18 }]\\ &= \left\{\;\exp\left[\beta\,\hbar\,\omega_j\right]-1\right\}^{-1}\delta_{jk}\end{aligned}$$

and Equation [VII-17] becomes

$$\begin{aligned}\left\langle F^{\dagger}(t)\,F(t')\right\rangle_{therm} &= \sum_{j}\left|\,g_j\,\right|^2\left\langle\, n_j\,\right\rangle_{therm}\exp\left[-i\left(\Omega-\omega_j\right)\left(t-t'\right)\right]\\ &= 2\,\pi\left|\,g(\Omega)\right|^2\rho\left(\Omega\right)\;\left\langle\, n(\Omega)\,\right\rangle_{therm}\delta\left(t-t'\right)\\ &= \gamma\left\langle\, n(\Omega)\,\right\rangle_{therm}\delta\left(t-t'\right) \qquad\qquad [\text{ VII-19 }]\\ &= 2\left\langle\, D_{A^{\dagger}A}\,\right\rangle_{therm}\delta\left(t-t'\right)\end{aligned}$$

where $\left\langle\, D_{A^{\dagger}A}\,\right\rangle_{therm} = \frac{\gamma}{4}\left\langle\, n\left(\Omega\right)\,\right\rangle_{therm}$ is of the form of a *diffusion coefficient* in the classical Langevin theory. Two-time correlation functions of this form are characteristic of **Markoffian** random processes and represent fluctuations in a reservoir with essentially zero memory. Substitution of this Markoffian correlation function into Equation [VII-16] yields an *Einstein relation* of the form

$$2\left\langle D_{A^\dagger A} \right\rangle_{therm} = \frac{d}{dt}\left\langle A^\dagger(t)\, A(t) \right\rangle_{therm} + \gamma \left\langle A^\dagger(t)\, A(t) \right\rangle_{therm} . \qquad [\text{ VII-20 }]$$

GENERAL RESEVOIR PROBLEM

As the previous section demonstrates, the effects of random fluctuations in an assemblage of harmonic oscillators may be accounted for in precise detail. The problem can be analyzed completely since the first term on the RHS of Equation [VII-6] -- the ***damping term*** -- contains only the system variable. In general, this feedback term will included reservoir variables as well and the simplified model breaks down. The general case may, however, be analyzed if we take the simpler case as a guide. In particular, let us suppose that a set of systems operators $\left\{ A_\mu(t) \right\}$ are coupled to the reservoir and satisfy a set Langevin equations of motion as

$$\dot{A}_\mu(t) = D_\mu(t) + F_\mu(t) \qquad [\text{ VII-21 }]$$

where $D_\mu(t)$ is the drift term and $F_\mu(t)$ is the Markoffian noise operator appropriate to the system operator $A_\mu(t)$. From the earlier results, we are, thus, assuming that the two-time correlation of the random fluctuating force is of the form

$$\left\langle F_\mu(t')\, F_\nu(t'') \right\rangle = 2\, D_{\mu\nu}\, \delta(t' - t'') \qquad [\text{ VII-22 }]$$

Again using the identity

$$A_\mu(t) = A_\mu(t - \Delta t) + \int_{t-\Delta t}^{t} dt'\, \dot{A}_\mu(t') \qquad [\text{ VII-23 }]$$

we find

$$\begin{aligned}\langle A_\mu(t)\, F_\nu(t)\rangle &= \langle A_\mu(t-\Delta t)\, F_\nu(t)\rangle \\ &\quad + \int_{t-\Delta t}^{t} dt'\,\langle D_\mu(t')\, F_\nu(t)\rangle + \int_{t-\Delta t}^{t} dt'\,\langle F_\mu(t')\, F_\nu(t)\rangle\end{aligned} \qquad [\text{ VII-24 }]$$

As discussed in connection with Equation [VII-14], the first and second term on the RHS in Equation [VII-24] vanishes since *cause* cannot precede *effect*. Therefore

$$\langle A_\mu(t)\, F_\nu(t)\rangle = \int_{t-\Delta t}^{t} dt'\,\langle F_\mu(t')\, F_\nu(t)\rangle \qquad [\text{ VII-25 }]$$

and by Equation [VII-22]

$$\langle A_\mu(t)\, F_\nu(t)\rangle = D_{\mu\,\nu} \qquad [\text{ VII-26 }]$$

We may then form the following equation of motion

$$\begin{aligned}\frac{d}{dt}\langle A_\mu\, A_\nu\rangle &= \langle \dot{A}_\mu\, A_\nu\rangle + \langle A_\mu\, \dot{A}_\nu\rangle \\ &= \langle D_\mu\, A_\nu\rangle + \langle F_\mu\, A_\nu\rangle + \langle A_\mu\, D_\nu\rangle + \langle A_\mu\, F_\nu\rangle\end{aligned} \qquad [\text{ VII-27 }]$$

which leads to a *generalized Einstein relationship*

$$2\,D_{\mu\,\nu} = \frac{d}{dt}\langle A_\mu\, A_\nu\rangle - \langle D_\mu\, A_\nu\rangle - \langle A_\mu\, D_\nu\rangle \qquad [\text{ VII-28 }]$$

With this equation it is possible to calculate the diffusion coefficients from the drift coefficients, provided one can independently calculate the equation of motion for $\langle A_\mu\, A_\nu\rangle$.

We can also use Equation [VII-21] to calculate the expectation values of two-time correlation functions. In paryicular, if multiply it by $A_\nu(t')$ where $t' < t$ and average we find

$$\frac{d}{dt}\left\langle A_\mu(t)\,A_\nu(t')\right\rangle = \left\langle D_\mu(t)\,A_\nu(t')\right\rangle + \left\langle F_\mu(t)\,A_\nu(t')\right\rangle \qquad \text{[VII-29]}$$

Since in the Markoff approximation, the present system operator $A_\nu(t')$ cannot know about the future noise source $F_\mu(t)$, $\left\langle F_\mu(t)\,A_\nu(t')\right\rangle$ vanishes and

$$\frac{d}{dt}\left\langle A_\mu(t)\,A_\nu(t')\right\rangle = \left\langle D_\mu(t)\,A_\nu(t')\right\rangle \qquad \text{[VII-30]}$$

which is the so called quantum regression theorm which states that the two-time correlation function $\left\langle A_\mu(t)\,A_\nu(t')\right\rangle$ satisfies the same equation of motion as the single-time $\left\langle A_\mu(t)\right\rangle$.

SEMICONDUCTOR LANGEVIN EQUATIONS

As discussed earlier, the effective Hamiltonian for optical interactions in a semiconductor may be written as

$$\mathcal{H}_{\text{eff}} = \sum_{\bar{\mathbf{k}}} \left\{ \left[\varepsilon_g + \frac{\hbar^2 k^2}{2m_{ec}} \right] a^{\dagger}_{\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}} + \left[\frac{\hbar^2 k^2}{2m_{hv}} \right] b^{\dagger}_{-\bar{\mathbf{k}}} b_{-\bar{\mathbf{k}}} - \left[g_{\bar{\mathbf{k}}} a^{\dagger}_{\bar{\mathbf{k}}} b^{\dagger}_{-\bar{\mathbf{k}}} a + g_{\bar{\mathbf{k}}} a^{\dagger} a_{\bar{\mathbf{k}}} b_{-\bar{\mathbf{k}}} \right] \right\} \qquad [\text{ VII-29 }]$$

Thus, the equation of motion motion for the effective dipole operator $b_{-\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}}$ is given by

$$\begin{aligned} \frac{d}{dt} b_{-\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}} &= \frac{i}{\hbar} \left[\mathcal{H}_{\text{eff}}, b_{-\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}} \right] \\ &= -\left(\gamma + i\,\omega_{\bar{\mathbf{k}}} \right) b_{-\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}} + i g_{\bar{\mathbf{k}}}\, a \left(a^{\dagger}_{\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}} + b^{\dagger}_{-\bar{\mathbf{k}}} b_{-\bar{\mathbf{k}}} - 1 \right) + f_{b_{-\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}}} \end{aligned} \qquad [\text{ VII-30 }]$$

where γ is the damping, $\omega_{\bar{\mathbf{k}}} = \varepsilon_g + \frac{\hbar^2 k^2}{2m_{ec}} + \frac{\hbar^2 k^2}{2m_{hv}}$, and $f_{b_{-\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}}}$ is the k-dependent noise operator. We transform the various operators to a frame rotating at the electromagnetic field frequency ω_r -- *viz.*

$$b_{-\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}} = \sigma_{\bar{\mathbf{k}}} \exp\left[-\omega_r\, t \right] \qquad [\text{ VII-31a }]$$

$$a(t) = A(t) \exp\left[-\omega_r\, t \right] \qquad [\text{ VII-31b }]$$

$$f_{b_{-\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}}} = f_{\sigma_{\bar{\mathbf{k}}}} = F_{\sigma_{\bar{\mathbf{k}}}} \exp\left[-\omega_r\, t \right] \qquad [\text{ VII-31c }]$$

and find the transformed equation of motion in the rotating frame -- *i.e.* the Heisenberg interaction picture -- as

$$\frac{d}{dt} \sigma_{\bar{\mathbf{k}}} = -\left(\gamma + i\,\omega_{\bar{\mathbf{k}}} - i\,\omega_r \right) \sigma_{\bar{\mathbf{k}}} + i g_{\bar{\mathbf{k}}} A \left(a^{\dagger}_{\bar{\mathbf{k}}} a_{\bar{\mathbf{k}}} + b^{\dagger}_{-\bar{\mathbf{k}}} b_{-\bar{\mathbf{k}}} - 1 \right) + F_{\sigma_{\bar{\mathbf{k}}}} \qquad [\text{ VII-32 }]$$

Applying the generalized Einstein relation -- *i.e.* Equation [VII-28] -- to the operator

$$\sigma^{\dagger}_{\vec{k}}\,\sigma_{\vec{k}} = a^{\dagger}_{\vec{k}}\, b^{\dagger}_{-\vec{k}}\, b_{-\vec{k}}\, a_{\vec{k}} = a^{\dagger}_{\vec{k}}\, a_{\vec{k}}\, b^{\dagger}_{-\vec{k}}\, b_{-\vec{k}} \equiv \sigma^{\vec{k}}_{33}$$

we obtain

$$\begin{aligned} 2\,D_{\sigma^{\dagger}_{\vec{k}}\,\sigma_{\vec{k}}} &= \left(\gamma + i\,\omega_{\vec{k}} - i\,\omega_r\right)\sigma^{\vec{k}}_{33} + \left(\gamma - i\,\omega_{\vec{k}} + i\,\omega_r\right)\sigma^{\vec{k}}_{33} + \sigma^{\vec{k}}_{33} \\ &\approx 2\,\gamma\,\sigma^{\vec{k}}_{33} \end{aligned} \qquad \text{[VII-33a]}$$

Similarly,

$$\begin{aligned} 2\,D_{\sigma_{\vec{k}}\,\sigma^{\dagger}_{\vec{k}}} &= \left(\gamma + i\,\omega_{\vec{k}} - i\,\omega_r\right)\sigma^{\vec{k}}_{00} + \left(\gamma - i\,\omega_{\vec{k}} + i\,\omega_r\right)\sigma^{\vec{k}}_{00} + \sigma^{\vec{k}}_{00} \\ &\approx 2\,\gamma\,\sigma^{\vec{k}}_{00} \end{aligned} \qquad \text{[VII-33b]}$$

where $\sigma^{\vec{k}}_{00} = \left(1 - a^{\dagger}_{\vec{k}}\, a_{\vec{k}}\right)\left(1 - b^{\dagger}_{-\vec{k}}\, b_{-\vec{k}}\right)$.

The equation of motion for the laser field operator in a frame rotating at the laser frequency ω_r may be written as

$$\dot{A}(t) = -\left[\frac{\omega}{2\,Q} + i\left(\Omega - \omega_r\right)\right]A(t) - i\sum_{\vec{k}} g_{\vec{k}}\,\sigma_{\vec{k}}(t) + F(t) \qquad \text{[VII-34]}$$

where Ω is the cold cavity resonant frequency and where we have replace the damping parameter γ in equation [VII-10] by the experimental parameter ω/Q . If we consider the Equation [VII-32] in the *quasi-equilbrium* limit, we find

$$\begin{aligned} \sigma_{\vec{k}}(t) &= \left[\gamma + i\,\omega_{\vec{k}} - i\,\omega_r\right]^{-1}\left[i g_{\vec{k}}\, A(t)\left(a^{\dagger}_{\vec{k}}\, a_{\vec{k}} + b^{\dagger}_{-\vec{k}}\, b_{-\vec{k}} - 1\right) + F_{\sigma_{\vec{k}}}(t)\right] \\ &= \mathcal{D}\left(\omega_{\vec{k}} - \omega_r ; \gamma\right)\left[i g_{\vec{k}}\, A(t)\left(\sigma^{\vec{k}}_{33} - \sigma^{\vec{k}}_{00}\right) + F_{\sigma_{\vec{k}}}(t)\right] \end{aligned} \qquad \text{[VII-35]}$$

where $\mathcal{D}(\omega_{\vec{k}} - \omega_r ; \gamma)$ is the complex Lorentzian demoninator.[45] Thus in the quasi-equilbrium limit, Equation [VII-34] becomes

$$\dot{A}(t) = -\left[\frac{\omega}{2\,Q} + i(\Omega - \omega_r) - \sum_{\vec{k}} \left|g_{\vec{k}}\right|^2 \mathcal{D}(\omega_{\vec{k}} - \omega_r ; \gamma)\left(\sigma_{33}^{\vec{k}} - \sigma_{00}^{\vec{k}}\right)\right] A(t) + F_A(t) \qquad \text{[VII-36]}$$

where

$$F_A(t) = F(t) - i\sum_{\vec{k}} g_{\vec{k}}\, \mathcal{D}(\omega_{\vec{k}} - \omega_r ; \gamma)\, F_{\sigma_{\vec{k}}}(t) \; . \qquad \text{[VII-37]}$$

Following arguments similar to those used in deriving Equation [VII-16], we easily derive an equation of motion for the laser photon number operator -- *viz*.

$$\begin{aligned} \frac{d}{dt}\left\langle A^{\dagger}(t)\, A(t)\right\rangle = &-\left[\frac{\omega}{Q} - \frac{2}{\gamma} \sum_{\vec{k}} \left|g_{\vec{k}}\right|^2 \mathcal{L}(\omega_{\vec{k}} - \omega_r ; \gamma)\left(\sigma_{33} - \sigma_{00}\right)\right]\left\langle A^{\dagger}(t)\, A(t)\right\rangle \\ &+ \int_{-\infty}^{+\infty} d\tau \left\langle F_A^{\dagger}(0), F_A(\tau)\right\rangle \end{aligned} \qquad \text{[VII-38a]}$$

or

$$\frac{d}{dt}\left\langle A^{\dagger}(t)\, A(t)\right\rangle = -\left[\frac{\omega}{Q} - \left(R_{33} - R_{00}\right)\right]\left\langle A^{\dagger}(t)\, A(t)\right\rangle + \int_{-\infty}^{+\infty} d\tau \left\langle F_A^{\dagger}(0), F_A(\tau)\right\rangle \qquad \text{[VII-38b]}$$

where

$$R_{33} = \frac{2}{\gamma}\sum_{\vec{k}} \left|g_{\vec{k}}\right|^2 \mathcal{L}(\omega_{\vec{k}} - \omega_r ; \gamma)\, \sigma_{33}^{\vec{k}} \; . \qquad \text{[VII-39a]}$$

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[45] It may be recalled that in the notes entitled *Lasers: Models and Theories* for convenience we indroduced the Lorentzian functions -- *viz*.

$$\mathcal{D}(u - v ; w) \equiv \left[i\,(u - v) + w\right]^{-1} \qquad \text{the } \textbf{complex Lorentzian denominator}$$

$$\mathcal{L}(u - v ; w) \equiv w^2\left[(u - v)^2 + w^2\right]^{-1} \qquad \text{the } \textbf{dimensionless Lorentzian function}.$$

and
$$R_{00} = \frac{2}{\gamma} \sum_{\vec{\mathbf{k}}} \left|g_{\vec{\mathbf{k}}}\right|^2 \mathcal{L}\left(\omega_{\vec{\mathbf{k}}} - \omega_r ; \gamma\right) \sigma_{00}^{\vec{\mathbf{k}}} \;. \qquad [\text{ VII-39b }]$$

Assuming that the cavity damping and carrier scattering reservoirs are uncorrelated, the two-time correlation function may be written as

$$\begin{aligned} \left\langle F_A^{\dagger}(t)\; F_A(t') \right\rangle &= \left\langle F^{\dagger}(t)\; F(t') \right\rangle \\ &\quad + \sum_{\vec{\mathbf{k}}} \sum_{\vec{\mathbf{k}}'} g_{\vec{\mathbf{k}}} g_{\vec{\mathbf{k}}'}\, \mathcal{D}\left(\omega_{\vec{\mathbf{k}}'} - \omega_r ; \gamma\right) \mathcal{D}\left(\omega_{\vec{\mathbf{k}}} - \omega_r ; \gamma\right) \left\langle F_{\sigma_{\vec{\mathbf{k}}}}^{\dagger}(t)\; F_{\sigma_{\vec{\mathbf{k}}'}}(t') \right\rangle \end{aligned} \qquad [\text{ VII-40a }]$$

and if the noise operators for different $\vec{\mathbf{k}}$ are uncorrelated

$$\left\langle F_A^{\dagger}(t)\; F_A(t') \right\rangle = \left\langle F^{\dagger}(t)\; F(t') \right\rangle + \frac{1}{\gamma^2} \sum_{\vec{\mathbf{k}}} \left|g_{\vec{\mathbf{k}}}\right|^2 \mathcal{L}\left(\omega_{\vec{\mathbf{k}}} - \omega_r ; \gamma\right) \left\langle F_{\sigma_{\vec{\mathbf{k}}}}^{\dagger}(t)\; F_{\sigma_{\vec{\mathbf{k}}'}}(t') \right\rangle \;. \qquad [\text{ VII-40b }]$$

Finally, using Equations [VII-19] and [VII-33a] we obtain

$$\left\langle F_A^{\dagger}(t)\; F_A(t') \right\rangle = \left[\frac{\omega}{Q} \left\langle n(\omega) \right\rangle_{therm} + \frac{2}{\gamma} \sum_{\vec{\mathbf{k}}} \left|g_{\vec{\mathbf{k}}}\right|^2 \mathcal{L}\left(\omega_{\vec{\mathbf{k}}} - \omega_r ; \gamma\right) \sigma_{33}^{\vec{\mathbf{k}}} \right] \delta(t - t') \;. \qquad [\text{ VII-40c }]$$

and, thus, the "$A^{\dagger} A$" diffusion coefficient for the laser field is

$$2\, D_{A^{\dagger} A} = \frac{\omega}{Q} \left\langle n(\omega) \right\rangle_{therm} + R_{33} \;. \qquad [\text{ VII-41a }]$$

By a similar argument, the "$A A^{\dagger}$" diffusion coefficient is

$$2\, D_{A A^{\dagger}} = \frac{\omega}{Q} \left[\left\langle n(\omega) \right\rangle_{therm} + 1 \right] + R_{00} \;. \qquad [\text{ VII-41b }]$$

Using Equation [VII-41a], Equation [VII-38b] becomes

$$\frac{d}{dt}\left\langle A^{\dagger}(t)\,A(t)\right\rangle = -\left[\frac{\omega}{Q} - \left(R_{33} - R_{00}\right)\right]\left\langle A^{\dagger}(t)\,A(t)\right\rangle + \frac{\omega}{Q}\,\left\langle n(\omega)\right\rangle_{therm} + R_{33} \qquad [\text{ VII-42a }]$$

Similarly

$$\frac{d}{dt}\left\langle A(t)\,A^{\dagger}(t)\right\rangle = -\left[\frac{\omega}{Q} - \left(R_{33} - R_{00}\right)\right]\left\langle A(t)\,A^{\dagger}(t)\right\rangle + \frac{\omega}{Q}\left[\left\langle n(\omega)\right\rangle_{therm} + 1\right] + R_{00} \qquad [\text{ VII-42b }]$$

As check on the consistency of these result, we see that the form of Equations [VII-42a] and [VII-42b] guarantees that the commutator $\left[A(t),\,A^{\dagger}(t)\right]$ is independent of time -- *viz*.

$$\frac{d}{dt}\left\langle\left[A(t),A^{\dagger}(t)\right]\right\rangle = -\left[\frac{\omega}{Q} - \left(R_{33} - R_{00}\right)\right]\left\langle\left[A(t),A^{\dagger}(t)\right]\right\rangle + \frac{\omega}{Q} + \left(R_{00} - R_{33}\right) = 0 \qquad [\text{ VII-43 }]$$

to incorporate saturation effects into the noise spectrum we need to write a Langevin equation for the carrier-density operator -- *viz*.

$$\begin{aligned}\frac{d}{dt}\,a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}} &= \frac{i}{\hbar}\left[\mathcal{H}_{\text{eff}},a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\right] + \left\{\text{damping, pumping, and noise terms}\right\}\\ &= \Lambda_{e}^{\vec{\mathbf{k}}}\left[1 - a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\right] - \Gamma_{\vec{\mathbf{k}}}\,a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\,b^{\dagger}_{-\vec{\mathbf{k}}}\,b_{-\vec{\mathbf{k}}} - \gamma_{nr}\,a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}} \qquad [\text{ VII-44 }]\\ &\quad - \gamma_{e}\left[a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}} - \left(a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\right)_{eq}\right] - i\left[g_{\vec{\mathbf{k}}}\,a^{\dagger}_{\vec{\mathbf{k}}}\,b^{\dagger}_{-\vec{\mathbf{k}}}\,a - g_{\vec{\mathbf{k}}}\,a^{\dagger}\,a_{\vec{\mathbf{k}}}\,b_{-\vec{\mathbf{k}}}\right] + F_{e}^{\vec{\mathbf{k}}}\end{aligned}$$

where $\Lambda_{e}^{\vec{\mathbf{k}}}\left[1 - a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\right]$ is the pumping due injected carriers, $\Gamma_{\vec{\mathbf{k}}}\,a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\,b^{\dagger}_{-\vec{\mathbf{k}}}\,b_{-\vec{\mathbf{k}}}$ is the radiative recombination (spontaneous emission) term, $\gamma_{nr}\,a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}$ is the nonradiative recombination term, $\gamma_{e}\left[a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\,b^{\dagger}_{-\vec{\mathbf{k}}} - \left(a^{\dagger}_{\vec{\mathbf{k}}}\,a_{\vec{\mathbf{k}}}\right)_{eq}\right]$ is the relaxation due carrier-carrier scattering, and $F_{e}^{\vec{\mathbf{k}}}$ is the carrier noise operator. In the quasi-equilibrium approximation -- *i.e.* from Equation [VII-35] -- we may sum Equation III-44] to obtain a Langevin equation for the total carrier-density operator

$$\begin{aligned}\frac{d}{dt}N = \Lambda - \frac{1}{V}\sum_{\vec{\mathbf{k}}}\Gamma_{\vec{\mathbf{k}}}\, a^{\dagger}_{\vec{\mathbf{k}}}\, a_{\vec{\mathbf{k}}}\, b^{\dagger}_{-\vec{\mathbf{k}}}\, b_{-\vec{\mathbf{k}}} - \gamma_{nr}\, N \\ - \frac{2\, A^{\dagger} A}{\gamma V}\sum_{\vec{\mathbf{k}}}\left|g_{\vec{\mathbf{k}}}\right|^{2}\mathcal{L}\left(\omega_{\vec{\mathbf{k}}} - \omega_{r};\gamma\right)\left(\sigma^{\vec{\mathbf{k}}}_{33} - \sigma^{\vec{\mathbf{k}}}_{00}\right) + F_{N}\end{aligned} \qquad \text{[VII-45a]}$$

where

$$F_{N}(t) = F_{e}^{\vec{\mathbf{k}}}(t) + \frac{i}{V}\sum_{\vec{\mathbf{k}}}\left[g_{\vec{\mathbf{k}}}\, A^{\dagger}(t)\,\mathcal{D}\left(\omega_{\vec{\mathbf{k}}} - \omega_{r};\gamma\right) F_{\sigma_{\vec{\mathbf{k}}}}(t) - g_{\vec{\mathbf{k}}}\, A(t)\,\mathcal{D}\left(\omega_{\vec{\mathbf{k}}} - \omega_{r};\gamma\right) F^{\dagger}_{\sigma_{\vec{\mathbf{k}}}}(t)\right] \qquad \text{[VII-45b]}$$

NOISE SPECTRA:

To calculate noise spectra we make use of semiclassical transformations -- *viz.*

$$A(t) = \left(E(t)/\mathcal{E}_{\omega_r}\right)\exp\left[-i\,\varphi(t)\right] \qquad \text{[VII-46a]}$$

$$A^{\dagger}(t) = \left(E(t)/\mathcal{E}_{\omega_r}\right)\exp\left[i\,\varphi(t)\right] \qquad \text{[VII-46b]}$$

$$E(t) = \mathcal{E}_{\omega_r}\sqrt{A^{\dagger}(t)\, A(t)} \qquad \text{[VII-46c]}$$

$$\varphi(t) = (i/2)\ \ln\left[A(t)/A^{\dagger}(t)\right] \qquad \text{[VII-46d]}$$

where $\mathcal{E}_{\omega_r} = \left[\hbar\,\omega_r/\varepsilon_0\, V\right]$ is the electric field per photon. Using the expression

$$D'_{\rho\sigma} = \sum_{\mu,\,\nu} D_{\mu\,\nu}\,\frac{\partial A'_{\rho}}{\partial A'_{\mu}}\,\frac{\partial A'_{\sigma}}{\partial A'_{\nu}} \qquad \text{[VII-47]}$$

we see that

$$\begin{aligned} D_{EE} &= D_{A^\dagger A}\,\frac{\partial E}{\partial A^\dagger}\,\frac{\partial E}{\partial A} + D_{A A^\dagger}\,\frac{\partial E}{\partial A}\,\frac{\partial E}{\partial A^\dagger} \\ &= \frac{\mathcal{E}_{\omega_r}^4}{4\,E^2}\left[A A^\dagger\, D_{A^\dagger A} + A^\dagger A\, D_{A A^\dagger}\right] \approx \frac{\mathcal{E}_{\omega_r}^4}{4}\left[D_{A^\dagger A} + D_{A A^\dagger}\right] \end{aligned} \qquad \text{[VII-48a]}$$

$$\begin{aligned} D_{\varphi\varphi} &= D_{A^\dagger A}\,\frac{\partial \varphi}{\partial A^\dagger}\,\frac{\partial \varphi}{\partial A} + D_{A A^\dagger}\,\frac{\partial \varphi}{\partial A}\,\frac{\partial \varphi}{\partial A^\dagger} \\ &= \frac{1}{4\,A A^\dagger}\,D_{A^\dagger A} + \frac{1}{4\,A^\dagger A}\,D_{A A^\dagger} \approx \frac{\mathcal{E}_{\omega_r}^2}{4\,E^2}\left[D_{A^\dagger A} + D_{A A^\dagger}\right] \end{aligned} \qquad \text{[VII-48a]}$$

VIII. NONLINEAR OPTICS -- QUANTUM PICTURE:[45]

A QUANTUM MECHANICAL VIEW OF THE BASICS OF NONLINEAR OPTICS[46]

In what follows we draw on the discussion of the density operator in Review of Basic Quantum Mechanics: Dynamic Behavior of Quantum Systems, Section II of the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory* (hereafter referred to as IRM:ST). The macroscopic polarization is given by

$$\left\langle \vec{\mathbf{P}} \right\rangle = \mathrm{Tr}\left(\rho\, \vec{\mathbf{P}} \right) \qquad [\text{ VIII-1 }]$$

We take the total Hamiltonian of a particular system in the form

$$\mathcal{H} \;=\; \mathcal{H}_0 + \mathcal{H}_{\mathrm{int}} + \mathcal{H}_{\mathrm{random}} \qquad [\text{ VIII-2 }]$$

where $\mathcal{H}_{\mathrm{random}}$ is a Hamiltonian describing the random perturbations on the system by the thermal reservoir surrounding the system. Thus

$$\frac{\partial \rho}{\partial t} = \frac{i}{\hbar}\left[\, \rho, \left(\mathcal{H}_0 + \mathcal{H}_{\mathrm{int}} \right) \right] + \left. \frac{\partial \rho}{\partial t} \right|_{\mathrm{relax}} \qquad [\text{ VIII-3 }]$$

where

$$\left. \frac{\partial \rho}{\partial t} \right|_{\mathrm{relax}} = \frac{i}{\hbar}\left[\, \rho, \mathcal{H}_{\mathrm{random}} \right] \qquad [\text{ VIII-4 }]$$

To find the nonlinear susceptibility, we make use of the following perturbation expansions:

$$\rho = \rho^{(0)} + \rho^{(1)} + \rho^{(2)} + \cdots \qquad [\text{ VIII-5a }]$$

---

[45] See Nonlinear Optics -- Classical Picture which is Section VII in the lecture set entitled *On Classical Electromagnetic Fields* (OCEF).

[46] See, for example, Chapter 2 in Y. R. Shen's *Principles of Nonlinear Optics*, Wiley (1984).

$$\left\langle \vec{\mathbf{P}} \right\rangle = \left\langle \vec{\mathbf{P}}^{(0)} \right\rangle + \left\langle \vec{\mathbf{P}}^{(1)} \right\rangle + \left\langle \vec{\mathbf{P}}^{(2)} \right\rangle + \cdots \qquad \text{[VIII-5b]}$$

with
$$\left\langle \vec{\mathbf{P}}^{(\alpha)} \right\rangle = \mathrm{Tr}\left(\rho^{(\alpha)} \, \vec{\mathbf{P}} \right) \qquad \text{[VIII-5c]}$$

By substituting these expansions into Equation [VIII-3] and equating terms of like order in $\mathcal{H}_{\text{int}}$, we obtain the following hierarchy of equations:

$$\frac{\partial \rho^{(0)}}{\partial t} = \frac{i}{\hbar}\left[\rho^{(0)}, \mathcal{H}_0 \right] + \left.\frac{\partial \rho^{(0)}}{\partial t}\right|_{\text{relax}} \qquad \text{[VIII-6a]}$$

$$\frac{\partial \rho^{(1)}}{\partial t} = \frac{i}{\hbar}\left\{ \left[\rho^{(1)}, \mathcal{H}_0 \right] + \left[\rho^{(0)}, \mathcal{H}_{\text{int}} \right] \right\} + \left.\frac{\partial \rho^{(1)}}{\partial t}\right|_{\text{relax}} \qquad \text{[VIII-6b]}$$

$$\frac{\partial \rho^{(2)}}{\partial t} = \frac{i}{\hbar}\left\{ \left[\rho^{(2)}, \mathcal{H}_0 \right] + \left[\rho^{(1)}, \mathcal{H}_{\text{int}} \right] \right\} + \left.\frac{\partial \rho^{(2)}}{\partial t}\right|_{\text{relax}} \qquad \text{[VIII-6c]}$$

$$\frac{\partial \rho^{(3)}}{\partial t} = \frac{i}{\hbar}\left\{ \left[\rho^{(3)}, \mathcal{H}_0 \right] + \left[\rho^{(2)}, \mathcal{H}_{\text{int}} \right] \right\} + \left.\frac{\partial \rho^{(3)}}{\partial t}\right|_{\text{relax}} \qquad \text{[VIII-6d]}$$

……………………………………………………

Following earlier considerations, it is reasonable to write

$$\left\langle n \right| \left.\frac{\partial \rho}{\partial t}\right|_{\text{relax}} \left| n' \right\rangle = -\Gamma_{nn'} \left\langle n \right| \rho \left| n' \right\rangle = -\Gamma_{nn'}\, \rho_{nn'} \qquad \text{[VIII-7]}$$

Since the perturbing field is resolvable into an **appropriate** set of Fourier components (either discrete or continuous), we may write

$$\mathcal{H}_{\text{int}} = \sum_k \mathcal{H}_{\text{int}}\left(\omega_k\right) \exp\left(-i\,\omega_k t\right) \qquad \text{[VIII-8a]}$$

$$\rho = \sum_k \rho\left(\omega_k\right) \exp\left(-i\,\omega_k t\right) \qquad \text{[VIII-8b]}$$

and resolve Equations [VIII-6] into a hierarchy of algebraic equations. The first member of that hierarchy becomes [47]

$$\begin{aligned} \rho^{(1)}_{nn'}\left(\omega_k\right)\left\{i\left(\omega_{nn'}-\omega_k\right)+\Gamma_{nn'}\right\} &= \frac{i}{\hbar}\left\langle n\right|\left\{\rho^{(0)}\,\mathcal{H}_{\text{int}}\left(\omega_k\right)-\mathcal{H}_{\text{int}}\left(\omega_k\right)\rho^{(0)}\right\}\left|n'\right\rangle \\ &= \frac{i}{\hbar}\left\langle n\right|\mathcal{H}_{\text{int}}\left(\omega_k\right)\left|n'\right\rangle\left\{\rho^{(0)}_{nn}-\rho^{(0)}_{n'n'}\right\} \end{aligned} \qquad \text{[VIII-9a]}$$

or[48]

$$\rho^{(1)}_{nn'}\left(\omega_k\right) = \frac{i}{\hbar}\left\langle n\right|\mathcal{H}_{\text{int}}\left(\omega_k\right)\left|n'\right\rangle\left\{\rho^{(0)}_{nn}-\rho^{(0)}_{n'n'}\right\}\mathcal{D}\left(\omega_{nn'}-\omega_k;\Gamma_{nn'}\right)\ . \qquad \text{[VIII-9b]}$$

The second member of the hierarchy becomes

$$\rho^{(2)}_{nn'}\left(\omega_k\right)\left\{\,i\left(\omega_{nn'}-\omega_k\right)+\Gamma_{nn'}\right\} = \frac{i}{\hbar}\sum_{k'}\left\langle n\right|\left[\,\rho^{(1)}\left(\omega_{k'}\right),\mathcal{H}_{\text{int}}\left(\omega_k-\omega_{k'}\right)\,\right]\left|n'\right\rangle \qquad \text{[VIII-10a]}$$

or

$$\begin{aligned} \rho^{(2)}_{nn'}\left(\omega_k\right) = \frac{i}{\hbar}\,\mathcal{D}\left(\omega_{nn'}-\omega_k;\Gamma_{nn'}\right)\sum_{k'}\sum_{n''}\Big\{\,&\rho^{(1)}_{nn''}\left(\omega_{k'}\right)\left\langle n''\right|\mathcal{H}_{\text{int}}\left(\omega_k-\omega_{k'}\right)\left|n'\right\rangle \\ &-\left\langle n\right|\mathcal{H}_{\text{int}}\left(\omega_k-\omega_{k'}\right)\left|n''\right\rangle\rho^{(1)}_{n''n'}\left(\omega_k\right)\Big\} \end{aligned} \qquad \text{[VIII-10b]}$$

---

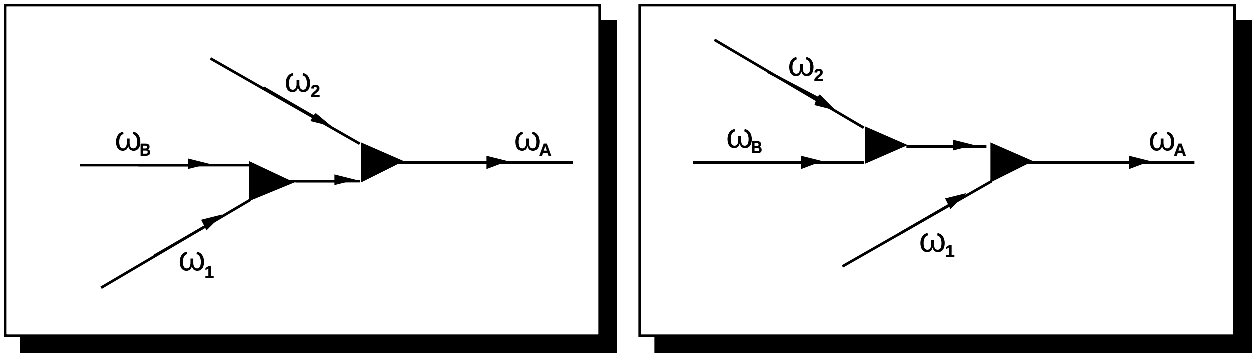
[47] we presume in this development that $\mathcal{H}_{\text{int}}$ is a Hermitian operator.

[48] Recall that $\mathcal{D}\left(u-v;\,w\right)\equiv\left[i\left(u-v\right)+w\right]^{-1}$ is the so called complex Lorentzian denominator.

Thus, we see that this expansion yields results akin to those embodied in Equations [VII-8] through [VIII-13] in OCEF.

TWO-PHOTON ABSORPTION

Let us consider two-photon absorption by a single atom (or by a set of paired $\vec{\mathbf{k}}$-states in a semiconductor as discussed Section 7 of IRM:ST).



Two-Photon Absorption Process

Tfrom earlier discussions, the appropriate interaction Hamiltonian is given by

$$\mathcal{H}_{2P} = -e\,\vec{\mathbf{r}}\;\vec{\mathbf{E}}^{(+)}\left(\vec{\mathbf{R}},t\right) = -e\,\vec{\mathbf{r}}\left[\vec{\mathbf{E}}_1^{(+)}\left(\vec{\mathbf{R}},t\right)+\vec{\mathbf{E}}_2^{(+)}\left(\vec{\mathbf{R}},t\right)\right] \qquad \text{[VIII-11]}$$

In the appendix to this section we establish -- *viz.*, in Equation [VIIIA-12] -- that the second approximation to the transition rate is given by

$$\frac{1}{\tau^{(2)}} \equiv \frac{d}{d\,t}\sum_f P_{i\to f}^{(2)}\left(t,t_0\right)$$

$$= \frac{2\pi}{\hbar^2}\sum_f \left|\left\langle f\right|\mathcal{H}_{\text{int}}\left|i\right\rangle + \frac{1}{\hbar}\sum_m \frac{\left\langle f\right|\mathcal{H}_{\text{int}}\left|m\right\rangle\left\langle m\right|\mathcal{H}_{\text{int}}\left|i\right\rangle}{\omega_i-\omega_m}\right|^2 \delta\left(\omega_i-\omega_f\right)$$

Thus for the two-photon process

$$\frac{1}{\tau^{(2)}} \qquad \frac{2\pi e^4}{\hbar^4}\sum_f \left|\sum_m \frac{\langle f|\vec{\mathbf{r}}\ \ \vec{\mathbf{E}}^{(+)}|m\rangle\langle m|\vec{\mathbf{r}}\ \ \vec{\mathbf{E}}^{(+)}|i\rangle}{\omega_i-\omega_m}\right|^2 \delta(\omega_i-\omega_f) \qquad \text{[VIII-12]}$$

For two beam, two-photon processes

$$\frac{1}{\tau^{(2)}} \qquad \frac{2\pi e^4}{\hbar^4}\sum_f \left|\sum_m \frac{\langle f|\vec{\mathbf{r}}\ \ \vec{\mathbf{E}}_2^{(+)}|m\rangle\langle m|\vec{\mathbf{r}}\ \ \vec{\mathbf{E}}_1^{(+)}|i\rangle}{\omega_1-\omega_m} + \frac{\langle f|\vec{\mathbf{r}}\ \ \vec{\mathbf{E}}_1^{(+)}|m\rangle\langle m|\vec{\mathbf{r}}\ \ \vec{\mathbf{E}}_2^{(+)}|i\rangle}{\omega_2-\omega_m}\right|^2 \delta(\omega_1+\omega_2-\omega_f) \qquad \text{[VIII-13a]}$$

and **for one beam, two-photon processes**

$$\frac{1}{\tau^{(2)}} \qquad \frac{2\pi e^4}{\hbar^4}\sum_f \left|\sum_m \frac{\langle f|\vec{\mathbf{r}}\ \ \vec{\mathbf{E}}^{(+)}|m\rangle\langle m|\vec{\mathbf{r}}\ \ \vec{\mathbf{E}}^{(+)}|i\rangle}{\omega_i-\omega_{\text{beam}}}\right|^2 \delta\left(2\omega_{\text{beam}}-\omega_f\right) \text{[VIII-13b]}$$

If we use once again the factorization used in Section VI of this lecture set -- *viz.* we write the initial state as $|\, i\,\rangle = |\, i = \{i', i''\}\rangle = |\, A\,(i')\,\rangle |\, F\,(i'')\,\rangle$ where $|\, A\,(i')\,\rangle$ and $|\, F\,(i'')\,\rangle$ are, respectively, the initial electronic (atomic) and field (photon) states. Using closure on the intermediate photon states, we find **for two beam, two-photon processes**

$$\frac{1}{\tau^{(2)}} \qquad \frac{2\pi e^4}{\hbar^4}\left|\mathcal{M}\ (1{,}2)\right|^2 \delta(\omega_1+\omega_2-\omega_f) \times \mathrm{Tr}\left[\rho\ \mathbf{E}_1^{(-)}\ \mathbf{E}_2^{(-)}\ \mathbf{E}_2^{(+)}\ \mathbf{E}_1^{(+)}\right] \qquad \text{[VIII-14a]}$$

where

$$\mathcal{M}\left(1,2\right) \equiv \sum_m \left[\frac{\left\langle A(f)\right| \vec{\mathbf{r}} \left|A(m)\right\rangle \ \varepsilon_1 \varepsilon_2 \ \left\langle A(m)\right| \vec{\mathbf{r}} \left|A(i)\right\rangle}{\omega_1 - \omega_m} + \frac{\left\langle A(f)\right| \vec{\mathbf{r}} \left|A(m)\right\rangle \ \hat{\varepsilon}_2 \hat{\varepsilon}_1 \ \left\langle A(m)\right| \vec{\mathbf{r}} \left|A(i)\right\rangle}{\omega_2 - \omega_m} \right] \qquad \text{[VIII-15a]}$$

and for **one beam, two-photon processes**

$$\frac{1}{\tau^{(2)}} \quad \frac{2\,\pi\,e^4}{\hbar^4} \left| \mathcal{M}\left(1,1\right) \right|^2 \delta\left(2\,\omega_1 - \omega_f\right) \times \mathrm{Tr}\left[\rho\, \mathbf{E}_1^{(-)}\ \mathbf{E}_1^{(-)}\ \mathbf{E}_1^{(+)}\ \mathbf{E}_1^{(+)}\right] \qquad \text{[VIII-14b]}$$

where

$$\mathcal{M}\left(1,1\right) \equiv \sum_m \frac{\left\langle A(f)\right| \vec{\mathbf{r}} \left|A(m)\right\rangle \ \varepsilon_1 \varepsilon_1 \ \left\langle A(m)\right| \vec{\mathbf{r}} \left|A(i)\right\rangle}{\omega_1 - \omega_m} \qquad \text{[VIII-15b]}$$

FOUR WAVE MIXING:

Four Wave Mixing Spectroscopy:

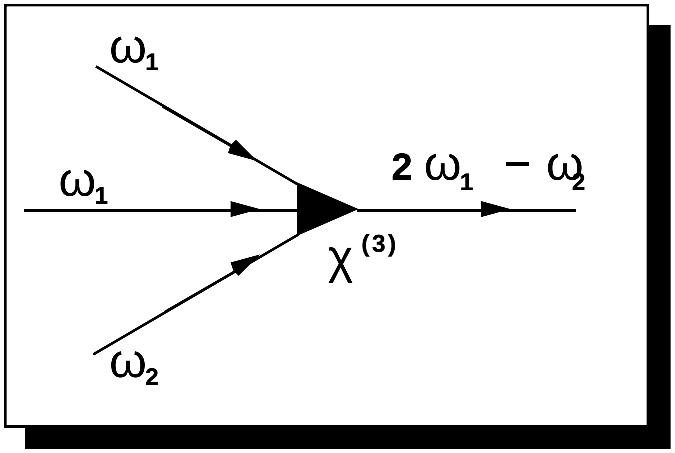
We explore here a particular set of four wave interactions. To that end, we consider an input field which consists of two plane waves

$$\mathbf{E}_\alpha\left(\vec{\mathbf{r}}, t\right) = \mathbf{E}_\alpha\left(\vec{\mathbf{k}}_1, \omega_1\right) \exp\left[i\left(\vec{\mathbf{k}}_1\ \vec{\mathbf{r}} - \omega_1\, t\right)\right] + \mathbf{E}_\alpha\left(\vec{\mathbf{k}}_2, \omega_2\right) \exp\left[i\left(\vec{\mathbf{k}}_2\ \vec{\mathbf{r}} - \omega_2 t\right)\right] + \text{c.c.} \qquad \text{[VIII-16]}$$

where frequencies ω_1 and ω_2 lie in the visible part of the spectrum. As Equation [VIII-3] of OCEF informs us, this input incident on a third-order nonlinear material, will **directly generate** a component of nonlinear polarization at a frequency $2\omega_1 - \omega_2$ -- *viz.*

$$
\begin{aligned}
{}^{(3)}\mathbf{P}_{\alpha}^{(NL)}\left(\vec{\mathbf{r}},t\right) = {} & 3\,\chi^{(3)}_{\alpha\beta\gamma\delta}\left(\omega_1;\omega_1;-\omega_2\right)\,\mathbf{E}_{\beta}\left(\vec{\mathbf{k}}_1,\,\omega_1\right)\mathbf{E}_{\gamma}\left(\vec{\mathbf{k}}_1,\,\omega_1\right)\mathbf{E}_{\delta}\left(\vec{\mathbf{k}}_2,\,\omega_2\right) \\
& \times \exp\left[i\left(2\vec{\mathbf{k}}_1-\vec{\mathbf{k}}_2\right)\,\vec{\mathbf{r}}\right]\exp\left[-i\left(2\omega_1-\omega_2\right)t\right]
\end{aligned}
\qquad [\text{ VIII-17 }]
$$

which may be associated with the destruction of two photons at ω_1 and the creation of photons at ω_2 and $2\omega_1-\omega_2$.



Four wave mixing - direct process

If, however, the material lacks a center of symmetry there is also an alternate path to obtain $2\omega_1-\omega_2$ radiation. In a second-order material [VIII-3] of OCEF informs us we can also have a nonlinear polarization at $2\omega_1-\omega_2$

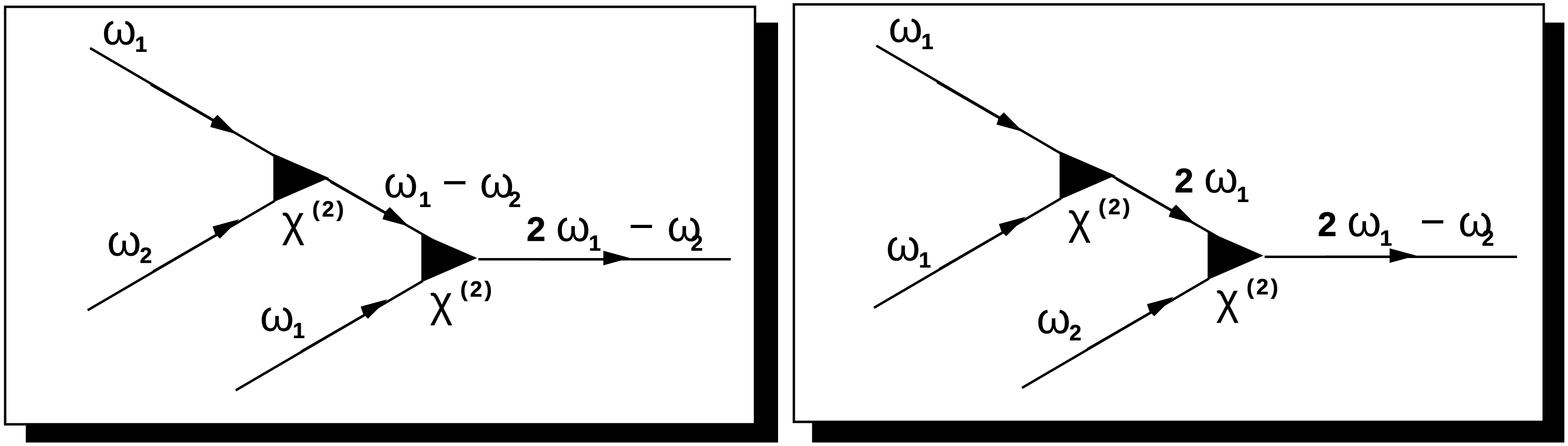
$$
\begin{aligned}
{}^{(2)}\mathbf{P}_{\alpha}^{(NL)}\left(\vec{\mathbf{r}},t\right) = {} & \left\{2\chi^{(2)}_{\alpha\beta\gamma}\left(\omega_1-\omega_2;\omega_1\right)\mathbf{E}_{\beta}\left(\vec{\mathbf{k}}_1-\vec{\mathbf{k}}_2,\,\omega_1-\omega_2\right)\mathbf{E}_{\gamma}\left(\vec{\mathbf{k}}_1,\,\omega_1\right)\right. \\
& \left.+\chi^{(2)}_{\alpha\beta\gamma}\left(2\omega_1;-\omega_2\right)\mathbf{E}_{\beta}\left(2\vec{\mathbf{k}}_1,\,2\omega_1\right)\mathbf{E}_{\gamma}\left(\vec{\mathbf{k}}_2,\,\omega_2\right)\right\} \\
& \times \exp\left[i\left(2\vec{\mathbf{k}}_1-\vec{\mathbf{k}}_2\right)\,\vec{\mathbf{r}}\right]\exp\left[-i\left(2\omega_1-\omega_2\right)t\right]
\end{aligned}
\qquad [\text{ VIII-18 }]
$$

if we have driving fields $\mathbf{E}_\beta\left(\vec{\mathbf{k}}_1-\vec{\mathbf{k}}_2,\ \omega_1-\omega_2\right)$ and $\mathbf{E}_\beta\left(2\vec{\mathbf{k}}_1,\ 2\omega_1\right)$. Such fields are, in fact, generate by the input field through the nonlinear polarization components

$$^{(2)}\mathbf{P}_\alpha^{(\mathrm{NL})}\left(\vec{\mathbf{k}}_1-\vec{\mathbf{k}}_2,\ \omega_1-\omega_2\right)=2\,\chi_{\alpha\beta\gamma}^{(2)}\left(\omega_1;-\omega_2\right)\mathbf{E}_\beta\left(\vec{\mathbf{k}}_1,\ \omega_1\right)\mathbf{E}_\gamma\left(\vec{\mathbf{k}}_2,\ \omega_2\right)\qquad[\text{ VIII-19a }]$$

and

$$^{(2)}\mathbf{P}_\alpha^{(\mathrm{NL})}\left(2\,\vec{\mathbf{k}}_1,\ 2\,\omega_1\right)=2\,\chi_{\alpha\beta\gamma}^{(2)}\left(\omega_1;\omega_1\right)\mathbf{E}_\beta\left(\vec{\mathbf{k}}_1,\ \omega_1\right)\mathbf{E}_\gamma\left(\vec{\mathbf{k}}_1,\ \omega_1\right)\qquad[\text{ VIII-19b }]$$



Four wave mixing - indirect processes

We may solve for $\mathbf{E}_\beta\left(\vec{\mathbf{k}}_1-\vec{\mathbf{k}}_2,\omega_1-\omega_2\right)$ and $\mathbf{E}_\beta\left(2\vec{\mathbf{k}}_1,\ 2\omega_1\right)$ from Equation [VIII-23] in OCEF -- *viz*.

$$\left\{\left|\vec{\mathbf{Q}}\right|^2\delta_{\alpha\beta}-\mathbf{Q}_\alpha\mathbf{Q}_\beta-\left(\omega^2/c^2\varepsilon_0\right)\varepsilon_{\alpha\beta}(\omega)\right\}\mathbf{E}_\beta\left(\vec{\mathbf{Q}},\ \omega\right)=\mu_0\,\omega^2\,\mathbf{P}_\alpha^{(\mathrm{NL})}\left(\vec{\mathbf{Q}},\omega\right)\qquad[\text{ VIII-20a }]$$

or using the operator $\overline{\overline{\;}}(\vec{\mathbf{k}})\equiv\left(\vec{\mathbf{k}}\ \ \vec{\mathbf{k}}\right)\overline{\overline{\mathbf{1}}}-\vec{\mathbf{k}}\,\vec{\mathbf{k}}=|k|^2\left\{\overline{\overline{\mathbf{1}}}-\breve{\mathbf{k}}\ \hat{\mathbf{k}}\right\}$ defined in OCEF

$$\left\{\varepsilon_0\,\varepsilon_{\alpha\beta}^{-1}(\omega)\ {}_{\beta\delta}\left(\vec{\mathbf{Q}}\right)-\left(\omega^2/c^2\right)\delta_{\alpha\delta}\right\}\mathbf{E}_\delta\left(\vec{\mathbf{Q}},\ \omega\right)=\left(\omega^2/c^2\right)\varepsilon_{\alpha\beta}^{-1}(\omega)\ \mathbf{P}_\beta^{(\mathrm{NL})}\left(\vec{\mathbf{Q}},\ \omega\right)\qquad[\text{ VIII-20b }]$$

If we define

$$\mathcal{G}_{\alpha\delta}\left(\vec{\mathbf{Q}}\,;\omega\right) \equiv \left\{ \varepsilon_0\, \varepsilon^{-1}_{\alpha\beta}(\omega) \quad {}_{\beta\delta}\left(\vec{\mathbf{Q}}\right) - \left(\omega^2/c^2\right)\delta_{\alpha\delta} \right\} \qquad \text{[VIII-21]}$$

we can, ***in principle***, write

$$\mathbf{E}_{\alpha}\left(\vec{\mathbf{Q}}, \omega\right) = \left[\omega^2/c^2\right] \mathcal{G}^{-1}_{\alpha\beta}\left(\vec{\mathbf{Q}}\,;\omega\right) \varepsilon^{-1}_{\beta\delta}(\omega)\, \mathbf{P}^{(NL)}_{\delta}\left(\vec{\mathbf{Q}}, \omega\right) \qquad \text{[VIII-22]}$$

Therefore, for a material lacking a center of symmetry, the complete nonlinear polarization may be written

$$\begin{aligned} \mathbf{P}^{(NL)}_{\alpha}\left(\vec{\mathbf{r}}, t\right) = {} & 3\exp\left[i\left(2\,\vec{\mathbf{k}}_1 - \vec{\mathbf{k}}_2\right)\ \vec{\mathbf{r}}\right] \exp\left[-i\left(2\,\omega_1 - \omega_2\right)\, t\right] \mathbf{E}_{\beta}\left(\vec{\mathbf{k}}_1, \omega_1\right) \mathbf{E}_{\gamma}\left(\vec{\mathbf{k}}_1, \omega_1\right) \mathbf{E}_{\delta}\left(\vec{\mathbf{k}}_2, \omega_2\right) \\ & \times\Big\{\chi^{(3)}_{\alpha\beta\gamma\delta}\left(\omega_1;\omega_1;-\omega_2\right) \\ & + \frac{4}{3}\left[\omega^2/c^2\right] \chi^{(2)}_{\alpha\eta\gamma}\left(\omega_1-\omega_2;\omega_1\right) \mathcal{G}^{-1}_{\eta\lambda}\left(\vec{\mathbf{k}}_1 - \vec{\mathbf{k}}_2, \omega_1 - \omega_2\right) \varepsilon^{-1}_{\lambda\varphi}\left(\omega_1-\omega_2\right) \chi^{(2)}_{\varphi\beta\delta}\left(\omega_1;-\omega_2\right) \\ & + \frac{2}{3}\left[\omega^2/c^2\right] \chi^{(2)}_{\alpha\eta\delta}\left(2\omega_1;-\omega_2\right) \mathcal{G}^{-1}_{\eta\lambda}\left(2\vec{\mathbf{k}}_1, 2\omega_1\right) \varepsilon^{-1}_{\lambda\varphi}\left(2\omega_1\right) \chi^{(2)}_{\varphi\beta\gamma}\left(\omega_1;\omega_1\right)\Big\} \end{aligned} \qquad \text{[VIII-23]}$$

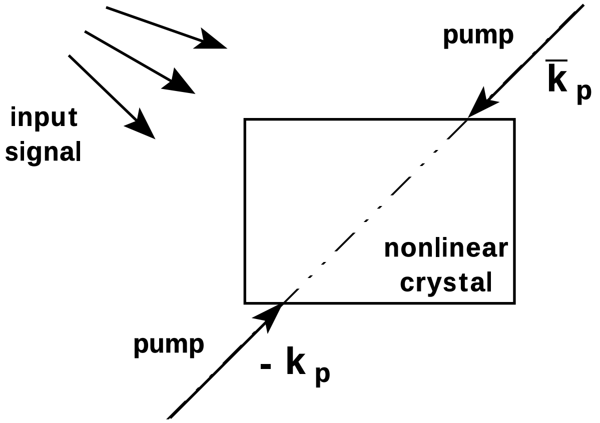
Notice the "resonance" in the second term when $\left|\vec{\mathbf{k}}_1 - \vec{\mathbf{k}}_2\right|$ and $\omega_1 - \omega_2$ are "tuned" to the wave number and frequency of coupled electromagnetic excitations or normal modes such as plasmons, polaritons and magnons.

Degenerate Four Wave Mixing (DFWM) - Phase Conjugation:

Consider now the reflected waves generated by the direct process -- *i.e.*, Equation [VIII-17] -- when the input field is the sum of two counter propagating "pump" fields, with $\left[\vec{\mathbf{k}}_p, \omega\right]$ and $\left[-\vec{\mathbf{k}}_p, \omega\right]$, plus a complex wave $\vec{\mathbf{E}}_{in}\left(\vec{\mathbf{r}}, t\right)$ represented a range of wave numbers components $\left[\left\{\vec{\mathbf{k}}_{\ell}\right\}, \omega\right]$ -- *viz.*

$$\begin{aligned}\mathbf{E}_\alpha(\vec{\mathbf{r}},t) = \Big\{ & \mathbf{E}_\alpha^{(+)} \exp\left[i\left(\vec{\mathbf{k}}_p \;\; \vec{\mathbf{r}} - \varphi_+\right)\right] + \mathbf{E}_\alpha^{(-)} \exp\left[i\left(-\vec{\mathbf{k}}_p \;\; \vec{\mathbf{r}} - \varphi_-\right)\right] \\ & + \sum_\ell \mathbf{E}_\alpha^{(\ell)} \exp\left[i\left(\vec{\mathbf{k}}_\ell \;\; \vec{\mathbf{r}} - \varphi_\ell\right)\right]\Big\} \exp\left[-i\,\omega\, t\right] + \text{c.c}\end{aligned} \qquad \text{[VIII-24]}$$

as shown below

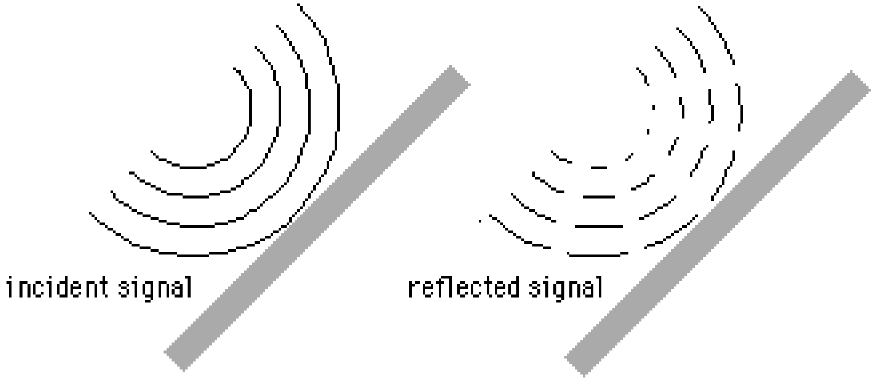


According to Equation [VIII-17], reflected waves are generated by nonlinear polarizations components with $\left[\left\{\vec{\mathbf{k}}_\ell\right\}, 3\omega\right]$, $\left[\left\{2\,\vec{\mathbf{k}}_p + \vec{\mathbf{k}}_\ell\right\}, \omega\right]$, $\left[\left\{2\,\vec{\mathbf{k}}_p - \vec{\mathbf{k}}_\ell\right\}, -\omega\right]$, and $\left[\left\{\vec{\mathbf{k}}_\ell\right\}, -\omega\right]$. The latter component is the so-called **phase conjugate term** and may be written

$$\begin{aligned}{}^{(3)}\mathbf{P}_\alpha^{(\mathrm{NL})}(\vec{\mathbf{r}},t) = \; & 3\chi^{(3)}_{\alpha\beta\gamma\delta}(\omega;\omega;\omega)\,\mathbf{E}_\beta^{(-)}\;\mathbf{E}_\gamma^{(+)}\left(\vec{\mathbf{k}}_p,\,\omega_1\right) \exp\left[-i\left(\varphi_+ + \varphi_-\right)\right] \\ & \sum_\ell \mathbf{E}_\delta^{(\ell)^*} \exp\left[-i\left(\vec{\mathbf{k}}_\ell \;\; \vec{\mathbf{r}} - \varphi_\ell\right)\right]\Big\} \exp\left[-i\,\omega\, t\right] + \text{c.c.}\end{aligned} \qquad \text{[VIII-25]}$$

which may be interpreted as the **time reversed image** of $\vec{\mathbf{E}}_{in}(\vec{\mathbf{r}},t)$.

TIME REVERSED IMAGING



THE RAMAN EFFECT

The Raman Nonlinearity:

In our discussion of the dielectric susceptibility thus far we have been implicitly assuming that the nuclei of system were clamped into fixed positions and have focused on the nonlinearities associate with electron dynamics. This is too limited a view. In general, we may expect that polarizability of, say, a molecule is a function of the nuclear positions and that the dipole moment can expressed

$$\mathbf{p}_\alpha\left(\left\{\vec{\mathbf{u}}^{(s)}\right\},\vec{\mathbf{E}}\right)=\mathbf{p}_\alpha^{(0)}\left(\left\{\vec{\mathbf{u}}^{(s)}\right\}\right)+a_{\alpha\beta}\left(\left\{\vec{\mathbf{u}}^{(s)}\right\}\right)\mathbf{E}_\beta+\cdots. \qquad [\text{ VIII-26 }]$$

$\left\{\vec{\mathbf{u}}^{(s)}\right\}$ denotes a collection of nuclear positions where $\vec{\mathbf{u}}^{(s)}$ is the displacement of the s nucleus from its nominal equilibrium position. Further, we may expand these terms in powers of the displacement -- *viz.*

$$\mathbf{p}_\alpha^{(0)}\left(\left\{\vec{\mathbf{u}}^{(i)}\right\}\right)\approx\mathbf{p}_\alpha^{(0)}+\sum_{s=1}^{N}\left[\frac{\partial}{\partial\mathbf{u}_\beta^{(s)}}\mathbf{p}_\alpha^{(0)}\left(\left\{\vec{\mathbf{u}}^{(s)}\right\}\right)\right]_0\mathbf{u}_\beta^{(s)}+\cdots \qquad [\text{ VIII-27a }]$$

$$\mathbf{p}_{\alpha}^{(1)}\left(\left\{\vec{\mathbf{u}}^{(i)}\right\},\vec{\mathbf{E}}\right)\equiv a_{\alpha\beta}\left(\left\{\vec{\mathbf{u}}^{(s)}\right\}\right)\mathbf{E}_{\beta}\approx a_{\alpha\beta}^{(0)}\;\mathbf{E}_{\beta}+\sum_{s=1}^{N}a_{\alpha\beta\gamma}^{(\mathrm{R})}\;\mathbf{E}_{\beta}\;\mathbf{u}_{\gamma}^{(s)}+\cdots \qquad [\text{ VIII-27b }]$$

The second term in Equation [VIII-27b] is the so called **Raman nonlinearity.** Since $\left[\frac{\partial}{\partial\,\mathbf{u}_{\beta}^{(s)}}\,p_{\alpha}^{(0)}\left(\left\{\vec{\mathbf{u}}^{(s)}\right\}\right)\right]_{0}$ has the dimensions of charge, Equation [VIII-27a] is usually written

$$\mathbf{p}_{\alpha}^{(0)}\left(\left\{\vec{\mathbf{u}}^{(s)}\right\}\right)\approx\mathbf{p}_{\alpha}^{(0)}+\sum_{s=1}^{N}e_{\alpha\beta}(s)\;\mathbf{u}_{\beta}^{(s)}+\cdots. \qquad [\text{ VIII-27a' }]$$

where $e_{\alpha\beta}(s)$ is the ***dynamic effective charge tensor*** associated with the s nucleus. The nuclear displacements are a linear combination of the normal modes of the system

$$\mathbf{u}_{\beta}^{(s)}=\sum_{\nu=1}^{3N}\Gamma_{\beta}(s;\nu)\;q(\nu) \qquad [\text{ VIII-28 }]$$

so that we may recast Equations [VIII-27a] and [VIII-27b] in the form

$$\mathbf{p}_{\alpha}^{(0)}\left(\left\{\vec{\mathbf{u}}^{(s)}\right\}\right)\approx p_{\alpha}^{(0)}+\sum_{\nu=1}^{3N}e_{\alpha}(\nu)\;q(\nu)+\cdots \qquad [\text{ VIII-29a }]$$

$$\mathbf{p}_{\alpha}^{(R)}=\sum_{s=1}^{N}a_{\alpha\beta\gamma}^{(\mathrm{R})}\;\mathbf{E}_{\beta}\;\mathbf{u}_{\gamma}^{(s)}=\sum_{\nu=1}^{3N}a_{\alpha\beta}^{(\mathrm{R})}(\nu)\;\mathbf{E}_{\beta}\;q(\nu) \qquad [\text{ VIII-29b }]$$

where $e_{\alpha}(\nu)=\sum_{s=1}^{N}e_{\alpha\beta}(s)\;\Gamma_{\beta}(s;\nu)$ and $a_{\alpha\beta}^{(\mathrm{R})}(\nu)=\sum_{s=1}^{N}a_{\alpha\beta\gamma}^{(\mathrm{R})}\;\Gamma_{\gamma}(s;\nu)$.

Quantum Theory of Raman Scattering

The Raman nonlinearity provides a coupling between the photon field described by the Hamiltonian

$$\mathcal{H}_{rad} = \sum_{\vec{\mathbf{k}},\sigma} \hbar\omega_{\vec{\mathbf{k}}} \left(a^{\dagger}_{\vec{\mathbf{k}},\sigma}\, a_{\vec{\mathbf{k}},\sigma} + \frac{1}{2} \right) \qquad \text{[VIII-30]}$$

and the phonon field described by the Hamiltonian

$$\mathcal{H}_{vib} = \sum_{\nu} \hbar\omega_{\nu} \left(b^{\dagger}_{\nu}\, b_{\nu} + \frac{1}{2} \right). \qquad \text{[VIII-31]}$$

where $b^{\dagger}_{\nu}$ and b_{ν} are, respectively, the phonon creation and destruction operators. In the electric dipole approximation, the interaction Hamiltonian can be written

$$\mathcal{H}_{\text{int}} = -\frac{1}{2} \sum_{\nu=1}^{3N} a^{(\mathrm{R})}_{\alpha\beta}(\nu)\, \mathbf{E}_{\alpha}\, \mathbf{E}_{\beta}\, q(\nu) \qquad \text{[VIII-32]}$$

In earlier discussions we have shown that

$$q(\nu) = \left(\frac{\hbar}{2M\,\omega_{\nu}} \right)^{1/2} \left(b^{\dagger}_{\nu} + b_{\nu} \right) \qquad \text{[VIII-33a]}$$

and

$$\mathbf{E}_{\alpha} = i \sum_{\vec{\mathbf{k}},\sigma} \left(\frac{2\pi\,\hbar\,\omega_{\vec{\mathbf{k}}}}{V} \right)^{1/2} \hat{\mathbf{e}}_{\alpha}\left(\vec{\mathbf{k}},\sigma\right) \left(a_{\vec{\mathbf{k}},\sigma} - a^{\dagger}_{\vec{\mathbf{k}},\sigma} \right) \qquad \text{[VIII-33b]}$$

so that the complete interaction Hamiltonian may expressed as

$$\mathcal{H}_{\text{int}} = \frac{1}{2}\left(\frac{2\pi\hbar}{V}\right)\sum_{\nu=1}^{3N}\left(\frac{\hbar}{2M\,\omega_\nu}\right)^{1/2} a_{\alpha\beta}^{(\mathrm{R})}(\nu)\left(b_\nu^\dagger + b_\nu\right)$$
$$\times\sum_{\vec{\mathbf{k}},\sigma}\omega_{\vec{\mathbf{k}}}^{1/2}\,\hat{\mathbf{e}}_\alpha\left(\vec{\mathbf{k}},\sigma\right)\left(a_{\vec{\mathbf{k}}\,\sigma} - a_{\vec{\mathbf{k}}\,\sigma}^\dagger\right)\sum_{\vec{\mathbf{k}}',\sigma'}\omega_{\vec{\mathbf{k}}'}^{1/2}\,\hat{\mathbf{e}}_\beta\left(\vec{\mathbf{k}}',\sigma'\right)\left(a_{\vec{\mathbf{k}}',\sigma'} - a_{\vec{\mathbf{k}}',\sigma'}^\dagger\right) \qquad [\text{ VIII-34 }]$$

To concentrate on the key issue, we write **scattering Hamiltonian** which is proportional to $a_{\vec{\mathbf{k}},\sigma}^\dagger\, a_{\vec{\mathbf{k}}',\sigma'}$ and which describes the scattering of a photon from the $\left\{\vec{\mathbf{k}}',\sigma'\right\}$ (initial) state to the $\left\{\vec{\mathbf{k}},\sigma\right\}$ (final) state

$$\mathcal{H}_{scat} = \left(\frac{2\pi\,\hbar}{V}\right)\left(\frac{\hbar}{2M\,\omega_\nu}\right)^{1/2}\left(\omega_I\,\omega_F\right)^{1/2} a^{(\mathrm{R})}(I,F;\nu)\, a_F^\dagger\, a_I\left(b_\nu^\dagger + b_\nu\right) \quad [\text{ VIII-35 }]$$

where $a^{(\mathrm{R})}(I,F;\nu) = a_{\alpha\beta}^{(\mathrm{R})}(\nu)\,\hat{\mathbf{e}}_\alpha(I)\,\hat{\mathbf{e}}_\beta(F)$.

We may now use the **Fermi golden rule**[49] to calculate the transition rate per "molecule" for **inelastic Stokes scattering** events -- *viz.* for $\left\{n_I, n_F, n_\nu\right\} \rightarrow \left\{n_I - 1, n_F + 1, n_\nu + 1\right\}$

$$\frac{1}{\tau^{(S)}} = \frac{2\pi}{\hbar^2}\left|\left\langle f\right|\mathcal{H}_{\text{scat}}\left|i\right\rangle\right|^2\delta\left(\omega_i - \omega_f\right)$$
$$= \frac{2\pi}{\hbar^2}\left(\frac{2\pi\,\hbar}{V}\right)^2\left(\frac{\hbar}{2M\,\omega_\nu}\right)\left(\omega_I\,\omega_F\right)\left|a^{(\mathrm{R})}(I,F;\nu)\right|^2 \qquad [\text{ VIII-36a }]$$
$$\times\left|\left\langle n_I - 1, n_F + 1, n_\nu + 1\right|\, a_F^\dagger\, a_I\, b_\nu^\dagger\left|n_I, n_F, n_\nu\right\rangle\right|^2\delta\left(\omega_I - \omega_F - \omega_\nu\right)$$

$$\frac{1}{\tau^{(S)}} = \frac{8\pi^3}{V^2}\left(\frac{\hbar}{2M\,\omega_\nu}\right)\left(\omega_I\,\omega_F\right)\left|\,a^{(\mathrm{R})}(I,F;\nu)\,\right|^2 n_I\left(n_F + 1\right)\left(n_\nu + 1\right)\delta\left(\omega_I - \omega_F - \omega_\nu\right) \quad [\text{ VIII-36b }]$$

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[49] See the appendix to this section for a recapitulation of the theory of transition rates.

and the transition rate per "molecule" for **inelastic anti-Stokes scattering** events -- *viz.* for $\left\{n_I, n_F, n_\nu\right\} \to \left\{n_I - 1, n_F + 1, n_\nu - 1\right\}$

$$\frac{1}{\tau^{(AS)}} = \frac{2\pi}{\hbar^2}\left(\frac{2\pi\hbar}{V}\right)^2\left(\frac{\hbar}{2M\,\omega_\nu}\right)(\omega_I\;\omega_F)\left|a^{(\mathrm{R})}(I,F;\nu)\right|^2$$

$$\times\left|\left\langle n_I - 1, n_F + 1, n_\nu - 1\right| a_F^\dagger\, a_I\, b_\nu \left|n_I, n_F, n_\nu\right\rangle\right|^2 \delta\left(\omega_I + \omega_\nu - \omega_F\right) \qquad [\text{ VIII-36c }]$$

$$= \frac{8\pi^3}{V^2}\left(\frac{\hbar}{2M\,\omega_\nu}\right)(\omega_I\;\omega_F)\left|a^{(\mathrm{R})}(I,F;\nu)\right|^2 n_I\,\left(n_F + 1\right) n_\nu\,\delta\left(\omega_I + \omega_\nu - \omega_F\right)$$

For a total number of "molecules" $\mathcal{V}\rho_{\mathrm{mol}}$ we may write the total time rate of change of photons of frequency ω_F as

$$\left.\frac{d\,n_F}{d\,t}\right|_S = \frac{8\pi^3\,\rho_{mol}\,\omega_I\,\omega_F}{V}\left(\frac{\hbar}{2M\,\omega_\nu}\right)\left|\,a^{(\mathrm{R})}(I,F;\nu)\,\right|^2$$

$$\times\, n_I\,\left(n_F + 1\right)\left(n_\nu + 1\right)\delta\left(\omega_I - \omega_F - \omega_\nu\right) \qquad [\text{ VIII-37a }]$$

$$\left.\frac{d\,n_F}{d\,t}\right|_{AS} = \frac{8\pi^3\,\rho_{mol}\,\omega_I\,\omega_F}{V}\left(\frac{\hbar}{2M\,\omega_\nu}\right)\left|\,a^{(\mathrm{R})}(I,F;\nu)\,\right|^2$$

$$\times\, n_I\,\left(n_F + 1\right) n_\nu\,\delta\left(\omega_I + \omega_\nu - \omega_F\right) \qquad [\text{ VIII-37b }]$$

Stimulated Raman Effect: Simple model

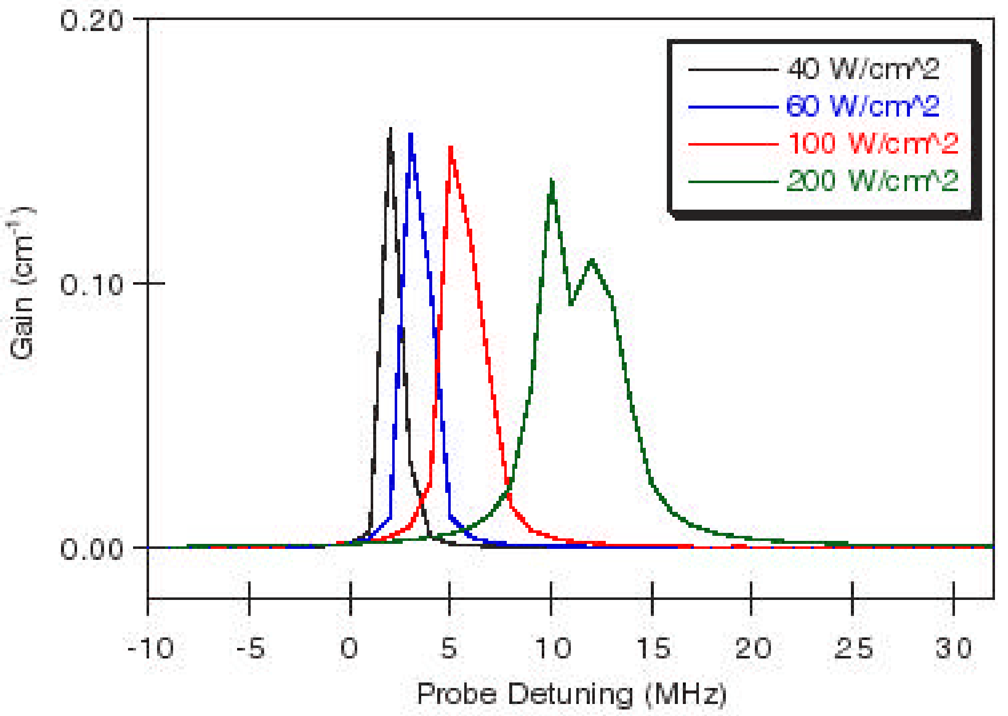
Suppose that a high power laser at ω_I -- **the pump --** propagates parallel to the z-axis and that the Raman medium occupies the half-space $z > 0$. The spatial rate of change of the *Stokes photons* at ω_F -- **the signal --** may be written[50]

$$\frac{d\,n_F(z)}{d\,z} = G_{\mathrm{R}}\;n_I(z)\left[n_F(z) + 1\right] \qquad [\text{ VIII-38 }]$$

---

[50] Assuming for the nonce that $n_\nu \approx 0$.

where
$$G_{\mathrm{R}} = \frac{8\pi^3\,\rho_{mol}\,\omega_I\,\omega_F}{cV\,\gamma_\nu}\left(\frac{\hbar}{2M\,\omega_\nu}\right)\left|a^{(\mathrm{R})}(I,F;\nu)\right|^2 \qquad [\text{ VIII-39 }]$$



Raman Gain in rubidium vapor for different coupling laser intensities

The rate constant γ_ν is included this expression for gain to account for the broadening of a given vibrational state. Each Stokes event, of course, also depletes the incident beam so that

$$\frac{d\,n_I(z)}{d\,z} = -G_{\mathrm{R}}\;n_I(z)\left[n_F(z)+1\right] \qquad [\text{ VIII-40 }]$$

which means that $$n_F(z) + n_I(z) = n_I(0) \ . \qquad \text{[VIII-41]}$$

Substituting this conservation condition into Equation [VIII-38] we see that

$$\frac{d\, n_F(z)}{d\, z} = G_{\mathrm{R}} \left[n_I(0) - n_F(z) \right] \left[n_F(z) + 1 \right] \qquad \text{[VIII-42a]}$$

or that $$\frac{d \approx n_F(z)}{\left[n_F(z) + 1 \right]} - \frac{d\, n_F(z)}{\left[n_F(z) - n_I(0) \right]} = G_{\mathrm{R}} \left[1 + n_I(0) \right] d\, z \qquad \text{[VIII-42b]}$$

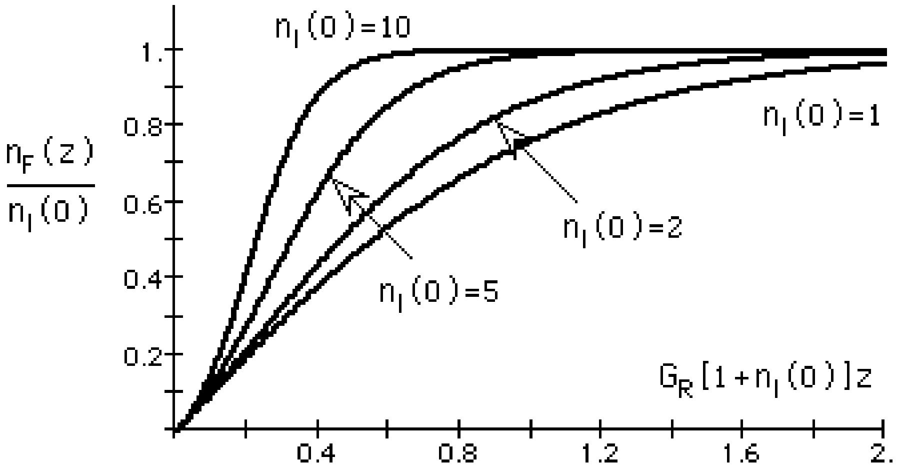
Therefore

$$n_F(z) = n_I(0) \, \frac{\exp\left\{ G_{\mathrm{R}} \left[1 + n_I(0) \right] z \right\} - 1}{n_I(0) + \exp\left\{ G_{\mathrm{R}} \left[1 + n_I(0) \right] z \right\}} \qquad \text{[VIII-43]}$$

This simple model predicts that Stokes photons initially increase linearly with distance and that there complete conversion at distances large compared to $\left\{ G_{\mathrm{R}} \left[1 + n_I(0) \right] \right\}^{-1}$ -- *i.e.*

$$\lim_{z \to 0} n_F(z) = G_{\mathrm{R}} \left[1 + n_I(0) \right] z \qquad \text{[VIII-44a]}$$

$$\lim_{z \to \infty} n_F(z) = n_I(0) \qquad \text{[VIII-44b]}$$



Coherent anti-Stokes Raman Spectroscopy (CARS):

There is an important four wave mixing process involving the Raman nonlinearity. We may see how this comes about by realizing that Equation [VIII-29] implies a potential energy contribution

$$\frac{1}{2}\sum_{\nu=1}^{3N} a_{\alpha\beta}^{(R)}(\nu)\,\mathbf{E}_\alpha\,\mathbf{E}_\beta\,q(\nu) \qquad \text{[VIII-45}$$

so that inclusion of the Raman nonlinearity modifies the equation of motion for the vibrational normal modes -- *viz.*

$$\ddot{q}(t,\nu)+\omega_\nu^2\,q(t,\nu)=\frac{1}{2M}\,a_{\alpha\beta}^{(R)}(\nu)\,\mathbf{E}_\alpha\,\mathbf{E}_\beta \qquad \text{[VIII-46]}$$

When the normal mode is driven by a two (visible) wave input (see discussion of four-wave processes above), its dominant response will be at the difference frequency $\omega_1-\omega_2$ -- *viz.*

$$\begin{aligned}\ddot{q}(t,\nu) &+ \omega_\nu^2\, q(t,\nu) \\ &= \frac{1}{M}\, a^{(R)}_{\alpha\beta}(\nu)\, \mathbf{E}_\alpha(\omega_1)\, E_\beta(\omega_2) \exp\left[-i\,(\omega_1-\omega_2)\, t\right] + c.c.\end{aligned} \qquad [\text{ VIII-47 }]$$

so that

$$q(t,\nu) = \frac{1}{M}\, \frac{a^{(R)}_{\alpha\beta}(\nu)\, E_\alpha(\omega_1)\, E_\beta(\omega_2)\, \exp\left[-i\,(\omega_1-\omega_2)\, t\right]}{\omega_\nu^2 - (\omega_1-\omega_2)^2} \qquad [\text{ VIII-48 }]$$

From Equation [VIII-29b] we see that excitation of a vibrational mode, through the Raman nonlinearity, generates a nonlinear polarization

$$\mathbf{p}^{(R)}_\alpha = \frac{1}{M} \sum_{\nu=1}^{3N} \frac{a^{(R)}_{\alpha\beta}(\nu)\, a^{(R)}_{\gamma\delta}(\nu)\, \mathbf{E}_\beta(\omega_1)\, \mathbf{E}_\gamma(\omega_1)\, \mathbf{E}_\delta(\omega_2)\, \exp\left[-i\,(2\omega_1-\omega_2)\, t\right]}{\omega_\nu^2 - (\omega_1-\omega_2)^2} \qquad [\text{ VIII-49 }]$$

at the four wave mixing frequency $2\omega_1 - \omega_2$. Thus, we see that the medium has an ***effective third order susceptibility***

$$\left[\, \chi^{(3)}_{\alpha\beta\gamma\delta}(2\,\omega_1 - \omega_2)\, \right]_{eff} = \chi^{(3)}_{\alpha\beta\gamma\delta} + \frac{1}{3M}\, \frac{a^{(R)}_{\alpha\beta}(\nu)\, a^{(R)}_{\gamma\delta}(\nu)}{\omega_\nu^2 - (\omega_1-\omega_2)^2} \qquad [\text{ VIII-50 }]$$

which exhibits a resonance when $\omega_1 - \omega_2$ equals ω_ν.

VIIIA. APPENDIX: RADIATIVE TRANSITION RATES REVISITED

This appendix builds on the formulation presented in Review of Basic Quantum Mechanics: Dynamic Behavior of Quantum Systems, Section II of the lecture notes entitled *The Interaction of Radiation and Matter: Semiclassical Theory* (hereafter referred to as IRM:ST) to obtain explicit and reasonably general expressions for radiative transition rates. Let us suppose that a coupled system of radiation and matter is described in the Schrödinger picture by a wave function $\left|\Psi_S(t)\right\rangle$. According to Equation [II-23] of IRM:ST, we may write

$$\left|\Psi_S(t)\right\rangle = \mathcal{T}_0(t)\left|\Psi_I(t)\right\rangle = \exp\left(-i\mathcal{H}_0\, t/\hbar\right)\left|\Psi_I(t)\right\rangle \qquad \text{[VIIIA-1]}$$

where $\mathcal{H}_0$ is the Hamiltonian of the uncoupled radiation and matter systems. Further, Equations [I-36a] and [I-37a] of IRM:ST inform us that

$$\left|\Psi_I(t)\right\rangle = \mathcal{T}_I(t,t_0)\left|\Psi_I(t_0)\right\rangle \qquad \text{[VIIIA-2]}$$

where

$$\begin{aligned} \mathcal{T}_I(t,t_0) &= 1 + \left(-\frac{i}{\hbar}\right)\int_{t_0}^{t} dt'\, \mathcal{H}^{I}_{\text{int}}(t')\, \mathcal{T}_I(t',t_0) \\ &= 1 + \left(-\frac{i}{\hbar}\right)\int_{t_0}^{t} dt'\, \mathcal{T}_0^{-1}(t')\, \mathcal{H}_{\text{int}}\, \mathcal{T}_0(t')\mathcal{T}_I(t',t_0) \end{aligned} \qquad \text{[VIIIA-3]}$$

In IRM:ST we showed how this integral equation can be iterated to yield $\mathcal{T}_I(t,t_0)$ as a power series in $\mathcal{H}^{I}_{\text{int}}(t)$. If denote the wave functions of the uncoupled system as $\left|\psi\right\rangle$, then the probability that the coupled system is in a state $\left|\psi_f\right\rangle$ at a time t is given by

$$\begin{aligned} \left|\langle\psi_f \,|\, \Psi_S(t)\,\rangle\right|^2 &= \left|\,\langle\psi_f|\mathcal{T}_0(t)|\Psi_I(t)\rangle\,\right|^2 \\ &= \left|\langle\psi_f|\mathcal{T}_0(t)\,\mathcal{T}_I(t,t_0)\,|\Psi_I(t_0)\rangle\right|^2 \qquad \text{[VIIIA-4]} \\ &= \left|\langle\psi_f|\mathcal{T}_0(t)\,\mathcal{T}_I(t,t_0)\,\mathcal{T}_0^{-1}(t_0)\,|\Psi_S(t_0)\rangle\right|^2 \end{aligned}$$

To be more specific, if the system happened to be in state $|\psi_i\rangle$ at a time t_0, the probability that the coupled system is in a state $|\psi_f\rangle$ at a time t is given by

$$P_{i\to f}(t,t_0) = \left|\langle\psi_f|\mathcal{T}_0(t)\,\mathcal{T}_I(t,t_0)\,\mathcal{T}_0^{-1}(t_0)\,|\psi_i\rangle\right|^2 \qquad \text{[VIIIA-5]}$$

In the most experimental circumstances, we are interested in the rate at which transitions take place from some particular initial state $|\psi_i\rangle$ to a set of final states $|\psi_f\rangle$ -- *i.e.*

$$\frac{1}{\tau} \equiv \frac{d}{d\,t}\sum_f P_{i\to f}\,(t,t_0) = \frac{d}{dt}\sum_f \left|\langle\psi_f|\mathcal{T}_0(t)\,\mathcal{T}_I(t,t_0)\,\mathcal{T}_0^{-1}(t_0)\,|\psi_i\rangle\right|^2 \qquad \text{[VIIIA-6]}$$

The first order approximation (for $1/\tau$):

Using the second term in the iteration set forth in Equation [II-27c] of IRM:ST, we can write

$$\begin{aligned} P^{(1)}_{i\to f}(t,t_0) &= \lim_{\varepsilon\to 0}\left|\langle\psi_f|\mathcal{T}_0(t)\left\{\left(-\frac{i}{\hbar}\right)\int_{-\infty}^{t} dt'\,\mathcal{T}_0^{-1}(t')\,\exp(\varepsilon\, t')\,\mathcal{H}_{\text{int}}\,\mathcal{T}_0(t')\right\}\mathcal{T}_0^{-1}(0)\,|\psi_i\rangle\right|^2 \\ &= \lim_{\varepsilon\to 0}\left|\left(-\frac{i}{\hbar}\right)\exp(-i\,\omega_f\,t)\,\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle\int_{-\infty}^{t} dt'\,\exp\left\{\left[i\,(\omega_f-\omega_i)+\varepsilon\right]\,t'\right\}\right|^2 \qquad \text{[VIIIA-7]} \\ &= \lim_{\varepsilon\to 0}\left|\frac{\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle}{\hbar}\;\frac{\exp(\varepsilon\, t - i\,\omega_i\,t)}{(\omega_i-\omega_f+i\,\varepsilon)}\right|^2 \end{aligned}$$

where the factor $\exp(\varepsilon\, t)$ was introduced to avoid the transient effects which might otherwise result from an apparent sudden application of the interaction between systems. Therefore, the first order approximation for $1/\tau$ is given by

$$\begin{aligned}
\frac{1}{\tau^{(1)}} &\equiv \frac{d}{d\,t}\sum_f P^{(1)}_{i\to f}(t,t_0) = \lim_{\varepsilon\to 0}\frac{d}{d\,t}\sum_f \left|\frac{\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle}{\hbar}\right|^2 \frac{\exp(2\varepsilon\, t)}{\left(\omega_i-\omega_f\right)^2+\varepsilon^2} \\
&= \lim_{\varepsilon\to 0}\frac{2}{\hbar^2}\sum_f \left|\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle\right|^2 \frac{\varepsilon\exp(2\varepsilon\, t)}{\left(\omega_i-\omega_f\right)^2+\varepsilon^2} \qquad \text{[VIIIA-8]} \\
&= \frac{2\pi}{\hbar^2}\sum_f \left|\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle\right|^2 \delta\left(\omega_i-\omega_f\right)
\end{aligned}$$

which is, of course, the generalized form of the **Fermi golden rule**.

The second order approximation (for $1/\tau$):

Using the third term in the iteration set forth in Equation [I-27c] of IRM:ST, we can write

$$\begin{aligned}
P^{(2)}_{i\to f}(t,t_0) = \lim_{\varepsilon\to 0}\Bigg| &\left(\frac{1}{\hbar}\right)\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle \frac{\exp(\varepsilon\, t - i\,\omega_i\, t)}{\left(\omega_i-\omega_f+i\,\varepsilon\right)} \\
&+\langle\psi_f|\mathcal{T}_0(t)\left\{\left(-\frac{i}{\hbar}\right)^2\int_{-\infty}^{t} dt'\,\exp(\varepsilon\, t')\,\mathcal{H}^{I}_{\text{int}}(t')\right. \qquad \text{[VIIIA-9a]} \\
&\left.\times\int_{-\infty}^{t'} dt''\,\exp(\varepsilon\, t'')\,\mathcal{H}^{I}_{1\text{int}}(t'')\right\}\mathcal{T}_0^{-1}(0)\,|\psi_i\rangle\Bigg|^2
\end{aligned}$$

or in the Schrödinger picture

$$P^{(2)}_{i\to f}(t,t_0) = \lim_{\varepsilon\to 0}\left|\left(\frac{1}{\hbar}\right)\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle\,\frac{\exp(\varepsilon\, t - i\,\omega_i\, t)}{(\omega_i-\omega_f+i\,\varepsilon)}\right.$$
$$+\langle\psi_f|\mathcal{T}_0(t)\left\{\left(-\frac{i}{\hbar}\right)^2\int_{-\infty}^{t}dt'\,\mathcal{T}_0^{-1}(t')\exp(\varepsilon\, t')\,\mathcal{H}_{\text{int}}\,\mathcal{T}_0(t')\right. \qquad \text{[VIIIA-9b]}$$
$$\left.\left.\times\int_{-\infty}^{t'}dt''\,\mathcal{T}_0^{-1}(t'')\exp(\varepsilon\, t'')\,\mathcal{H}_{\text{int}}\,\mathcal{T}_0(t'')\right\}\mathcal{T}_0^{-1}(0)\,|\psi_i\rangle\right|^2$$

Using the closure theorem we see that

$$P^{(2)}_{i\to f}(t,t_0) = \lim_{\varepsilon\to 0}\left|\left(\frac{1}{\hbar}\right)\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle\,\frac{\exp(\varepsilon\, t - i\,\omega_i\, t)}{(\omega_i-\omega_f+i\,\varepsilon)}\right.$$
$$+\left(-\frac{i}{\hbar}\right)^2\sum_m \exp(-i\,\omega_f\, t)\,\langle\psi_f|\,\mathcal{H}_{\text{int}}\,|\psi_m\rangle\langle\psi_m|\,\mathcal{H}_{\text{int}}\,|\psi_i\rangle \qquad \text{[VIIIA-10]}$$
$$\left.\times\int_{-\infty}^{t}dt'\int_{-\infty}^{t'}dt''\,\exp\left[i\,\omega_f\, t'+\varepsilon\, t'-i\,\omega_m\,(t'-t'')+\varepsilon\, t''-i\,\omega_i\, t''\right]\right|^2$$

Integrating and writing the matrix elements in more concise notation, we see that

$$P^{(2)}_{i\to f}(t,t_0) = \lim_{\varepsilon\to 0}\left|\frac{\exp(\varepsilon\, t-i\,\omega_i\, t)}{(\omega_i-\omega_f+i\,\varepsilon)}\left\{\left(\frac{1}{\hbar}\right)\langle f|\,\mathcal{H}_{\text{int}}\,|i\rangle\right.\right.$$
$$\left.\left.+\left(\frac{1}{\hbar}\right)^2\sum_m\frac{\langle f|\,\mathcal{H}_{\text{int}}\,|m\rangle\langle m|\,\mathcal{H}_{\text{int}}\,|i\rangle}{\omega_i-\omega_m+i\,\varepsilon/2}\right\}\right|^2 \qquad \text{[VIIIA-11]}$$

Therefore, following the arguments presented above, we see that the second order approximation for $1/\tau$ is given by

$$\begin{aligned}\frac{1}{\tau^{(2)}} &\equiv \frac{d}{d\,t}\sum_f P^{(2)}_{i\to f}\left(t,t_0\right)\\ &= \frac{2\pi}{\hbar^2}\sum_f\left|\langle f|\,\mathcal{H}_{\text{int}}\,|i\rangle+\frac{1}{\hbar}\sum_m\frac{\langle f|\,\mathcal{H}_{\text{int}}\,|m\rangle\langle m|\,\mathcal{H}_{\text{int}}\,|i\rangle}{\omega_i-\omega_m}\right|^2\delta\left(\omega_i-\omega_f\right)\end{aligned}\tag*{[VIIIA-12]}$$

The nth order approximation (for $1/\tau$):

We then may make the obvious extrapolation of these results and write the nth order approximation for $1/\tau$ as

$$\begin{aligned}\frac{1}{\tau^{(n)}} = \frac{2\pi}{\hbar^2}\sum_f\Bigg|&\langle f|\,\mathcal{H}_{\text{int}}\,|i\rangle+\frac{1}{\hbar}\sum_{m_1}\frac{\langle f|\,\mathcal{H}_{\text{int}}\,|m_1\rangle\langle m_1|\,\mathcal{H}_{\text{int}}\,|i\rangle}{\omega_i-\omega_{m_1}}+\dots\\ &+\left(\frac{1}{\hbar}\right)^{n-1}\sum_{m_1}\sum_{m_2}\dots\sum_{m_{n-1}}\frac{\langle f|\,\mathcal{H}_{\text{int}}\,|m_1\rangle\langle m_1|\,\mathcal{H}_{\text{int}}\,|m_2\rangle\dots\langle m_{n-1}|\,\mathcal{H}_{\text{int}}\,|i\rangle}{\left(\omega_i-\omega_{m_1}\right)\left(\omega_i-\omega_{m_2}\right)\dots\left(\omega_i-\omega_{m_{n-1}}\right)}\Bigg|^2\delta\left(\omega_i-\omega_f\right)\end{aligned}\tag*{[VIIIA-13]}$$

where $|m_1\rangle$, $|m_2\rangle$, *etc.* could be real imtermediate states or so called ***virtual intermediate states*** for the transitions to the state $|f\rangle$. The conservation of energy holds only between $|i\rangle$ and $|f\rangle$.